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# Mathematical analysis of plasmonic resonances for nanoparticles: the full Maxwell equations\*

Habib Ammari<sup>†</sup>    Matias Ruiz<sup>‡</sup>    Sanghyeon Yu<sup>†</sup>    Hai Zhang<sup>§</sup>

## Abstract

In this paper we use the full Maxwell equations for light propagation in order to analyze plasmonic resonances for nanoparticles. We mathematically define the notion of plasmonic resonance and analyze its shift and broadening with respect to changes in size, shape, and arrangement of the nanoparticles, using the layer potential techniques associated with the full Maxwell equations. We present an effective medium theory for resonant plasmonic systems and derive a condition on the volume fraction under which the Maxwell-Garnett theory is valid at plasmonic resonances.

Mathematics Subject Classification (MSC2000): 35R30, 35C20.

Keywords: plasmonic resonance, Neumann-Poincaré operator, nanoparticle, scattering and absorption enhancements, Maxwell equations, Maxwell-Garnett theory.

## 1 Introduction

The aim of this paper is to analyze plasmon resonant nanoparticles. Plasmon resonant nanoparticles have unique capabilities of enhancing the brightness and directivity of light, confining strong electromagnetic fields, and outcoupling of light into advantageous directions [44]. Recent advances in nanofabrication techniques have made it possible to construct complex nanostructures such as arrays using plasmonic nanoparticles as components. A thriving interest for optical studies of plasmon resonant nanoparticles is due to their recently proposed use as labels in molecular biology [28]. New types of cancer diagnostic nanoparticles are constantly being developed. Nanoparticles are also being used in thermotherapy as nanometric heat-generators that can be activated remotely by external electromagnetic fields [19]. Plasmon resonances in nanoparticles can be treated at the quasi-static limit as an eigenvalue problem for the Neumann-Poincaré integral operator [6, 26, 38, 39]. At this limit, they are size-independent. However, as the particle size increases, they are determined from scattering and absorption blow up and become size-dependent. This was experimentally observed, for instance, in [45].

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The objective of this paper is twofold: (i) To analytically investigate the plasmonic resonances of a single nanoparticle and analyze the shift and broadening of the plasmon resonance with changes in size and shape of the nanoparticles using the full Maxwell equations; (ii) To derive a Maxwell-Garnett type theory for approximating the plasmonic resonances of a periodic arrangement of nanoparticles. The paper generalizes to the full Maxwell equations the results obtained in [12, 17] where the Helmholtz equation was used to model light propagation. It provides the first mathematical study of the shift in plasmon resonance using the full Maxwell equations. On the other hand, it rigorously shows the validity of the Maxwell-Garnett theory for arbitrary-shaped nanoparticles at plasmonic resonances. The paper is organized as follows. In section 2 we first review commonly used function spaces. Then we introduce layer potentials associated with the Laplace operator and recall their mapping properties. In section 3 we first derive a layer potential formulation for the scattering problem and then we obtain a first-order correction to plasmonic resonances in terms of the size of the nanoparticle. This will enable us to analyze the shift and broadening of the plasmon resonance with changes in size and shape of the nanoparticles. The resonance condition is determined from absorption and scattering blow up and depends on the shape, size and electromagnetic parameters of both the nanoparticle and the surrounding material. Surprisingly, it turns out that in this case not only the spectrum of the Neumann-Poincaré operator plays a role in the resonance of the nanoparticles, but also its negative. We explain how in the quasi-static limit, only the spectrum of the Neumann-Poincaré operator can be excited. However, when the particle size increases and deviates from the dipole approximation, the resonances become size-dependent. Moreover, a part of the spectrum of negative of the Neumann-Poincaré operator can be excited as in higher-order terms in the expansion of the electric field versus the size of the particle. In section 4 we establish the quasi-static limit for the electromagnetic fields and derive a formula for the enhancement of the extinction cross-section. It is not clear for what kind of geometries in  $\mathbb{R}^3$  the spectrum of the Neumann-Poincaré operator has symmetries, that is, if  $\lambda \in \sigma(\mathcal{K}_D^*)$  so does  $-\lambda$ . In section 5 we provide calculations for the case of spherical nanoparticles wherein these symmetries are not present and we explicitly compute the shift in the spectrum of the Neumann-Poincaré operator and the extinction cross-section. In section 6 we consider the case of a spherical shell and apply degenerate perturbation theory since the eigenvalues associated with the corresponding Neumann-Poincaré operator are not simple. It is also worth mentioning that the spectrum of the associated Neumann-Poincaré operator is symmetric around zero. In section 7 we analyze the anisotropic quasi-static problem in terms of layer potentials and define the plasmonic resonances for anisotropic nanoparticles. Formulas for a small anisotropic perturbation of resonances of the isotropic formulas are derived. Finally, section 8 is devoted to establish a Maxwell-Garnett type theory for approximating the plasmonic resonances of a periodic arrangement of arbitrary-shaped nanoparticles. The Maxwell-Garnett theory provides a simple model for calculating the macroscopic optical properties of materials with a dilute inclusion of spherical nanoparticles [9]. It is widely used to assign effective properties to systems of nanoparticles. We rigorously obtain effective properties of a periodic arrangement of arbitrary-shaped nanoparticles and derive a condition on the volume fraction of the nanoparticles that insures the validity of the Maxwell-Garnett theory for predicting the effective optical properties of systems of embedded in a dielectric host material at the plasmonic resonances.

## 2 Preliminaries

Let us first fix some notation, definitions and recall some useful results for the rest of this paper.

- For a simply connected domain  $D \Subset \mathbb{R}^3$ ,  $\nu$  denotes the outward normal to  $\partial D$  and  $\frac{\partial}{\partial \nu}$  the outward normal derivative;
- $\varphi|_{\pm}(x) = \lim_{t \rightarrow 0^+} \varphi(x \pm t\nu)$ ;
- $Id$  denotes the identity operator;
- $\nabla \times$  denotes the curl operator for a vector field in  $\mathbb{R}^3$ ;
- For any functional space  $E(\partial D)$  defined on  $\partial D$ ,  $E_0(\partial D)$  denotes its zero mean subspace.

Here and throughout this paper, we assume that  $D$  is simply connected and of class  $\mathcal{C}^{1,\alpha}$  for  $0 < \alpha < 1$ .

Let  $H^s(\partial D)$  denote the usual Sobolev space of order  $s$  on  $\partial D$  and

$$H_T^s(\partial D) = \left\{ \varphi \in (H^s(\partial D))^3, \nu \cdot \varphi = 0 \right\}.$$

Let  $\nabla_{\partial D}$ ,  $\nabla_{\partial D} \cdot$  and  $\Delta_{\partial D}$  denote the surface gradient, surface divergence and Laplace-Beltrami operator respectively and define the vectorial and scalar surface curl by  $\text{curl}_{\partial D} \varphi = -\nu \times \nabla_{\partial D} \varphi$  for  $\varphi \in H^{\frac{1}{2}}(\partial D)$  and  $\text{curl}_{\partial D} \varphi = -\nu \cdot (\nabla_{\partial D} \times \varphi)$  for  $\varphi \in H_T^{-\frac{1}{2}}(\partial D)$ , respectively. Remind that

$$\begin{aligned} \nabla_{\partial D} \cdot \nabla_{\partial D} &= \Delta_{\partial D}, \\ \text{curl}_{\partial D} \vec{\text{curl}}_{\partial D} &= -\Delta_{\partial D}, \\ \nabla_{\partial D} \cdot \vec{\text{curl}}_{\partial D} &= 0, \\ \text{curl}_{\partial D} \nabla_{\partial D} &= 0. \end{aligned}$$

We introduce the following functional space:

$$H_T^{-\frac{1}{2}}(\text{div}, \partial D) = \left\{ \varphi \in H_T^{-\frac{1}{2}}(\partial D), \nabla_{\partial D} \cdot \varphi \in H^{-\frac{1}{2}}(\partial D) \right\}.$$

Let  $G$  be the Green function for the Helmholtz operator  $\Delta + k^2$  satisfying the Sommerfeld radiation condition in dimension three

$$\left| \frac{\partial G}{\partial |x|} - ikG \right| \leq C|x|^{-2}$$

for some constant  $C$  as  $|x| \rightarrow +\infty$ , uniformly in  $x/|x|$ .

The Green function  $G$  is given by

$$G(x, y, k) = -\frac{e^{ik|x-y|}}{4\pi|x-y|}. \quad (2.1)$$

Define the following boundary integral operators

$$\begin{aligned}\vec{\mathcal{S}}_D^k[\varphi] : H_T^{-\frac{1}{2}}(\partial D) &\longrightarrow H_T^{\frac{1}{2}}(\partial D) \\ \varphi &\longmapsto \vec{\mathcal{S}}_D^k[\varphi](x) = \int_{\partial D} G(x, y, k) \varphi(y) d\sigma(y), \quad x \in \mathbb{R}^3;\end{aligned}\tag{2.2}$$

$$\begin{aligned}\mathcal{S}_D^k[\varphi] : H^{-\frac{1}{2}}(\partial D) &\longrightarrow H^{\frac{1}{2}}(\partial D) \\ \varphi &\longmapsto \mathcal{S}_D^k[\varphi](x) = \int_{\partial D} G(x, y, k) \varphi(y) d\sigma(y), \quad x \in \mathbb{R}^3;\end{aligned}\tag{2.3}$$

$$\begin{aligned}\mathcal{K}_D^*[\varphi] : H^{-\frac{1}{2}}(\partial D) &\longrightarrow H^{-\frac{1}{2}}(\partial D) \\ \varphi &\longmapsto \mathcal{K}_D^*[\varphi](x) = \int_{\partial D} \frac{\partial G(x, y, 0)}{\partial \nu(x)} \varphi(y) d\sigma(y), \quad x \in \partial D;\end{aligned}\tag{2.4}$$

$$\begin{aligned}\mathcal{M}_D^k[\varphi] : H_T^{-\frac{1}{2}}(\text{div}, \partial D) &\longrightarrow H_T^{-\frac{1}{2}}(\text{div}, \partial D) \\ \varphi &\longmapsto \mathcal{M}_D^k[\varphi](x) = \int_{\partial D} \nu(x) \times \nabla_x \times G(x, y, k) \varphi(y) d\sigma(y), \quad x \in \partial D;\end{aligned}\tag{2.5}$$

$$\begin{aligned}\mathcal{L}_D^k[\varphi] : H_T^{-\frac{1}{2}}(\text{div}, \partial D) &\longrightarrow H_T^{-\frac{1}{2}}(\text{div}, \partial D) \\ \varphi &\longmapsto \mathcal{L}_D^k[\varphi](x) = \nu(x) \times \left( k^2 \vec{\mathcal{S}}_D^k[\varphi](x) + \nabla \mathcal{S}_D^k[\nabla_{\partial D} \cdot \varphi](x) \right), \quad x \in \partial D.\end{aligned}\tag{2.6}$$

Throughout this paper, we denote  $\vec{\mathcal{S}}_D^0, \mathcal{S}_D^0, \mathcal{M}_D^0$  by  $\vec{\mathcal{S}}_D, \mathcal{S}_D, \mathcal{M}_D$ , respectively. We also denote  $\mathcal{K}_D$  by the  $(\cdot, \cdot)_{-\frac{1}{2}, \frac{1}{2}}$ -adjoint of  $\mathcal{K}_D^*$ , where  $(\cdot, \cdot)_{-\frac{1}{2}, \frac{1}{2}}$  is the duality pairing between  $H^{-\frac{1}{2}}(\partial D)$ .

We recall now some useful results on the operator  $\mathcal{K}_D^*$  [7, 16, 32, 34].

**Lemma 2.1.** (i) *The following Calderón identity holds:  $\mathcal{K}_D \mathcal{S}_D = \mathcal{S}_D \mathcal{K}_D^*$ ;*

(ii) *The operator  $\mathcal{K}_D^*$  is compact self-adjoint in the Hilbert space  $H^{-\frac{1}{2}}(\partial D)$  equipped with the following inner product*

$$(u, v)_{\mathcal{H}^*} = -(u, \mathcal{S}_D[v])_{-\frac{1}{2}, \frac{1}{2}},\tag{2.7}$$

*which is equivalent to  $(\cdot, \cdot)_{-\frac{1}{2}, \frac{1}{2}}$ ;*

(iii) *Let  $(\lambda_j, \varphi_j)$ ,  $j = 0, 1, 2, \dots$  be the eigenvalue and normalized eigenfunction pair of  $\mathcal{K}_D^*$  in  $\mathcal{H}^*(\partial D)$ . Then,  $\lambda_j \in (-\frac{1}{2}, \frac{1}{2}]$ ,  $\lambda_j \neq 1/2$  for  $j \geq 1$ ,  $\lambda_j \rightarrow 0$  as  $j \rightarrow \infty$  and  $\varphi_j \in \mathcal{H}_0^*(\partial D)$  for  $j \geq 1$ , where  $\mathcal{H}_0^*(\partial D)$  is the zero mean subspace of  $\mathcal{H}^*(\partial D)$ ;*

(iv) *The following representation formula holds: for any  $\psi \in H^{-1/2}(\partial D)$ ,*

$$\mathcal{K}_D^*[\psi] = \sum_{j=0}^{\infty} \lambda_j (\psi, \varphi_j)_{\mathcal{H}^*} \otimes \varphi_j;$$

(v) The following trace formula holds: for any  $\psi \in \mathcal{H}^*(\partial D)$ ,

$$(\pm \frac{1}{2}Id + \mathcal{K}_D^*)[\varphi] = \frac{\partial \mathcal{S}_D[\varphi]}{\partial \nu} \Big|_{\pm}.$$

(vi) Let  $\mathcal{H}(\partial D)$  be the space  $H^{\frac{1}{2}}(\partial D)$  equipped with the following equivalent inner product

$$(u, v)_{\mathcal{H}} = -(\mathcal{S}_D^{-1}[u], v)_{-\frac{1}{2}, \frac{1}{2}}. \quad (2.8)$$

Then,  $\mathcal{S}_D$  is an isometry between  $\mathcal{H}^*(\partial D)$  and  $\mathcal{H}(\partial D)$ .

The following result holds.

**Lemma 2.2.** *The following Helmholtz decomposition holds [25]:*

$$H_T^{-\frac{1}{2}}(\text{div}, \partial D) = \nabla_{\partial D} H^{\frac{3}{2}}(\partial D) \oplus \vec{\text{curl}}_{\partial D} H^{\frac{1}{2}}(\partial D).$$

**Remark 2.1.** *The Laplace-Beltrami operator  $\Delta_{\partial D} : H_0^{\frac{3}{2}}(\partial D) \rightarrow H_0^{-\frac{1}{2}}(\partial D)$  is invertible. Here  $H_0^{\frac{3}{2}}(\partial D)$  and  $H_0^{-\frac{1}{2}}(\partial D)$  are the zero mean subspaces of  $H^{\frac{3}{2}}(\partial D)$  and  $H^{-\frac{1}{2}}(\partial D)$  respectively.*

The following results on the operator  $\mathcal{M}_D$  are of great importance.

**Lemma 2.3.**  $\mathcal{M}_D : H_T^{-\frac{1}{2}}(\text{div}, \partial D) \longrightarrow H_T^{-\frac{1}{2}}(\text{div}, \partial D)$  is a compact operator.

**Lemma 2.4.** *The following identities hold [6, 27]:*

$$\begin{aligned} \mathcal{M}_D[\vec{\text{curl}}_{\partial D} \varphi] &= \vec{\text{curl}}_{\partial D} \mathcal{K}_D[\varphi], \quad \forall \varphi \in H^{\frac{1}{2}}(\partial D), \\ \mathcal{M}_D[\nabla_{\partial D} \varphi] &= -\nabla_{\partial D} \Delta_{\partial D}^{-1} \mathcal{K}_D^*[\Delta_{\partial D} \varphi] + \vec{\text{curl}}_{\partial D} \mathcal{R}_D[\varphi], \quad \forall \varphi \in H^{\frac{3}{2}}(\partial D), \end{aligned}$$

where  $\mathcal{R}_D = -\Delta_{\partial D}^{-1} \vec{\text{curl}}_{\partial D} \mathcal{M}_D \nabla_{\partial D}$ .

### 3 Layer potential formulation for the scattering problem

We consider the scattering problem of a time-harmonic electromagnetic wave incident on a plasmonic nanoparticle. The homogeneous medium is characterized by electric permittivity  $\varepsilon_m$  and magnetic permeability  $\mu_m$ , while the particle occupying a bounded and simply connected domain  $D \subseteq \mathbb{R}^3$  of class  $\mathcal{C}^{1,\alpha}$  for  $0 < \alpha < 1$  is characterized by electric permittivity  $\varepsilon_c$  and magnetic permeability  $\mu_c$ , both of which depend on the frequency. Define

$$k_m = \omega \sqrt{\varepsilon_m \mu_m}, \quad k_c = \omega \sqrt{\varepsilon_c \mu_c},$$

and

$$\varepsilon_D = \varepsilon_m \chi(\mathbb{R}^3 \setminus \bar{D}) + \varepsilon_c \chi(D), \quad \mu_D = \mu_m \chi(\mathbb{R}^3 \setminus \bar{D}) + \mu_c \chi(D),$$

where  $\chi$  denotes the characteristic function.

For a given incident plane wave  $(E^i, H^i)$ , solution to the Maxwell equations in free space

$$\begin{aligned} \nabla \times E^i &= i\omega \mu_m H^i \quad \text{in } \mathbb{R}^3, \\ \nabla \times H^i &= -i\omega \varepsilon_m E^i \quad \text{in } \mathbb{R}^3, \end{aligned}$$

the scattering problem can be modeled by the following system of equations

$$\begin{aligned} \nabla \times E &= i\omega\mu_D H \quad \text{in } \mathbb{R}^3 \setminus \partial D, \\ \nabla \times H &= -i\omega\varepsilon_D E \quad \text{in } \mathbb{R}^3 \setminus \partial D, \\ \nu \times E|_+ - \nu \times E|_- &= \nu \times H|_+ - \nu \times H|_- = 0 \quad \text{on } \partial D, \end{aligned} \quad (3.1)$$

subject to the Silver-Müller radiation condition:

$$\lim_{|x| \rightarrow \infty} |x| \left( \sqrt{\mu_m} (H - H^i)(x) \times \frac{x}{|x|} - \sqrt{\varepsilon_m} (E - E^i)(x) \right) = 0$$

uniformly in  $x/|x|$ . Here and throughout the paper, the subscripts  $\pm$  indicate, as said before, the limits from outside and inside  $D$ , respectively.

Using the boundary integral operators (2.2) and (2.5), the solution to (3.1) can be represented as [46]

$$E(x) = \begin{cases} E^i(x) + \mu_m \nabla \times \bar{\mathcal{S}}_D^{k_m}[\psi](x) + \nabla \times \nabla \times \bar{\mathcal{S}}_D^{k_m}[\phi](x) & x \in \mathbb{R}^3 \setminus \bar{D}, \\ \mu_c \nabla \times \bar{\mathcal{S}}_D^{k_c}[\psi](x) + \nabla \times \nabla \times \bar{\mathcal{S}}_D^{k_c}[\phi](x) & x \in D, \end{cases} \quad (3.2)$$

and

$$H(x) = -\frac{i}{\omega\mu_D} (\nabla \times E)(x) \quad x \in \mathbb{R}^3 \setminus \partial D, \quad (3.3)$$

where the pair  $(\psi, \phi) \in (H_T^{-\frac{1}{2}}(\text{div}, \partial D))^2$  is the unique solution to

$$\begin{pmatrix} \frac{\mu_c + \mu_m}{2} Id + \mu_c \mathcal{M}_D^{k_c} - \mu_m \mathcal{M}_D^{k_m} & \mathcal{L}_D^{k_c} - \mathcal{L}_D^{k_m} \\ \mathcal{L}_D^{k_c} - \mathcal{L}_D^{k_m} & \left( \frac{k_c^2}{2\mu_c} + \frac{k_m^2}{2\mu_m} \right) Id + \frac{k_c^2}{\mu_c} \mathcal{M}_D^{k_c} - \frac{k_m^2}{\mu_m} \mathcal{M}_D^{k_m} \end{pmatrix} \begin{pmatrix} \psi \\ \phi \end{pmatrix} = \begin{pmatrix} \nu \times E^i \\ i\omega\nu \times H^i \end{pmatrix} \Big|_{\partial D}. \quad (3.4)$$

Let  $D = z + \delta B$  where  $B$  contains the origin and  $|B| = O(1)$ . For any  $x \in \partial D$ , let  $\tilde{x} = \frac{x-z}{\delta} \in \partial B$  and define for each function  $f$  defined on  $\partial D$ , a corresponding function defined on  $B$  as follows

$$\eta(f)(\tilde{x}) = f(z + \delta\tilde{x}). \quad (3.5)$$

Throughout this paper, for two Banach spaces  $X$  and  $Y$ , by  $\mathcal{L}(X, Y)$  we denote the set of bounded linear operators from  $X$  into  $Y$ . We will also denote by  $\mathcal{L}(X)$  the set  $\mathcal{L}(X, X)$ .

**Lemma 3.1.** *For  $\varphi \in H_T^{-\frac{1}{2}}(\text{div}, \partial D)$ , the following asymptotic expansion holds*

$$\mathcal{M}_D^k[\varphi](x) = \mathcal{M}_B[\eta(\varphi)](\tilde{x}) + \sum_{j=2}^{\infty} \delta^j \mathcal{M}_{B,j}^k[\eta(\varphi)](\tilde{x}),$$

where

$$\mathcal{M}_{B,j}^k[\eta(\varphi)](\tilde{x}) = \int_{\partial B} \frac{-(ik)^j}{4\pi j!} \nu(\tilde{x}) \times \nabla_{\tilde{x}} \times |\tilde{x} - \tilde{y}|^{j-1} \eta(\varphi)(\tilde{y}) d\sigma(\tilde{y}).$$

Moreover,  $\|\mathcal{M}_{B,j}^k\|_{\mathcal{L}(H_T^{-\frac{1}{2}}(\text{div}, \partial B))}$  is uniformly bounded with respect to  $j$ . In particular, the

convergence holds in  $\mathcal{L}(H_T^{-\frac{1}{2}}(\text{div}, \partial B))$  and  $\mathcal{M}_D^k$  is analytical in  $\delta$ .

*Proof.* We can see, after a change of variables, that

$$\mathcal{M}_D^k[\varphi](x) = \int_{\partial B} \nu(\tilde{x}) \times \nabla_{\tilde{x}} \times G(\tilde{x}, \tilde{y}, \delta k) \eta(\varphi)(\tilde{y}) d\sigma(\tilde{y}).$$

A Taylor expansion of  $G(\tilde{x}, \tilde{y}, \delta k)$  yields

$$G(\tilde{x}, \tilde{y}, \delta k) = - \sum_{j=0}^{\infty} \frac{(i\delta k |\tilde{x} - \tilde{y}|)^j}{j! 4\pi |\tilde{x} - \tilde{y}|} = - \frac{1}{4\pi |\tilde{x} - \tilde{y}|} + \sum_{j=1}^{\infty} \delta^j \frac{(ik)^j}{4\pi j!} |\tilde{x} - \tilde{y}|^{j-1},$$

hence

$$\mathcal{M}_D^k[\varphi](x) = \mathcal{M}_B[\eta(\varphi)](\tilde{x}) + \sum_{j=2}^{\infty} \delta^j \int_{\partial B} \frac{-(ik)^j}{4\pi j!} \nu(\tilde{x}) \times \nabla_{\tilde{x}} \times |\tilde{x} - \tilde{y}|^{j-1} \eta(\varphi)(\tilde{y}) d\sigma(\tilde{y}).$$

where it is clear from the regularity of  $|\tilde{x} - \tilde{y}|^{j-1}$ ,  $j \geq 2$ , that  $\|\mathcal{M}_{B,j}^k[\eta(\varphi)]\|_{H_T^{-\frac{1}{2}}(\text{div}, \partial B)}$  is uniformly bounded with respect to  $j$ , therefore,  $\|\mathcal{M}_{B,j}^k\|_{\mathcal{L}(H_T^{-\frac{1}{2}}(\text{div}, \partial B))}$  is uniformly bounded with respect to  $j$  as well.  $\square$

**Lemma 3.2.** For  $\varphi \in H_T^{-\frac{1}{2}}(\text{div}, \partial D)$ , the following asymptotic expansion holds

$$(\mathcal{L}_D^{k_c} - \mathcal{L}_D^{k_m})[\varphi](x) = \sum_{j=1}^{\infty} \delta^j \omega \mathcal{L}_{B,j}[\eta(\varphi)](\tilde{x}),$$

where

$$\mathcal{L}_{B,j}[\eta(\varphi)](\tilde{x}) = C_j \nu(\tilde{x}) \times \left( \int_{\partial B} |\tilde{x} - \tilde{y}|^{j-2} \eta(\varphi)(\tilde{y}) d\sigma(\tilde{y}) - \int_{\partial B} \frac{|\tilde{x} - \tilde{y}|^{j-2} (\tilde{x} - \tilde{y})}{j+1} \nabla_{\partial B} \cdot \eta(\varphi)(\tilde{y}) d\sigma(\tilde{y}) \right),$$

and

$$C_j = \frac{i^j (k_c^{j+1} - k_m^{j+1})}{\omega 4\pi (j-1)!}.$$

Moreover,  $\|\mathcal{L}_{B,j}\|_{\mathcal{L}(H_T^{-\frac{1}{2}}(\text{div}, \partial B))}$  is uniformly bounded with respect to  $j$ . In particular, the convergence holds in  $\mathcal{L}(H_T^{-\frac{1}{2}}(\text{div}, \partial B))$  and  $\mathcal{L}_D^k$  is analytical in  $\delta$ .

*Proof.* The proof is similar to that of Lemma 3.1.  $\square$

Using Lemma 3.1 and Lemma 3.2, we can write the system of equations (3.4) as follows:

$$\mathcal{W}_B(\delta) \begin{pmatrix} \eta(\psi) \\ \omega \eta(\phi) \end{pmatrix} = \left( \begin{array}{c} \frac{\eta(\nu \times E^i)}{\mu_m - \mu_c} \\ \frac{\eta(i\nu \times H^i)}{\varepsilon_m - \varepsilon_c} \end{array} \right) \Big|_{\partial B}, \quad (3.6)$$



where

$$\mathcal{W}_B(\delta) = \begin{pmatrix} \lambda_\mu Id - \mathcal{M}_B + \delta^2 \frac{\mu_m \mathcal{M}_{B,2}^{k_m} - \mu_c \mathcal{M}_{B,2}^{k_c}}{\mu_m - \mu_c} + O(\delta^3) & \frac{1}{\mu_m - \mu_c} (\delta \mathcal{L}_{B,1} + \delta^2 \mathcal{L}_{B,2}) + O(\delta^3) \\ \frac{1}{\varepsilon_m - \varepsilon_c} (\delta \mathcal{L}_{B,1} + \delta^2 \mathcal{L}_{B,2}) + O(\delta^3) & \lambda_\varepsilon Id - \mathcal{M}_B + \delta^2 \frac{\varepsilon_m \mathcal{M}_{B,2}^{k_m} - \varepsilon_c \mathcal{M}_{B,2}^{k_c}}{\varepsilon_m - \varepsilon_c} + O(\delta^3) \end{pmatrix}, \quad (3.7)$$

and

$$\lambda_\mu = \frac{\mu_c + \mu_m}{2(\mu_m - \mu_c)}, \quad \lambda_\varepsilon = \frac{\varepsilon_c + \varepsilon_m}{2(\varepsilon_m - \varepsilon_c)}. \quad (3.8)$$

It is clear that

$$\mathcal{W}_B(0) = \mathcal{W}_{B,0} = \begin{pmatrix} \lambda_\mu Id - \mathcal{M}_B & 0 \\ 0 & \lambda_\varepsilon Id - \mathcal{M}_B \end{pmatrix}.$$

Moreover,

$$\mathcal{W}_B(\delta) = \mathcal{W}_{B,0} + \delta \mathcal{W}_{B,1} + \delta^2 \mathcal{W}_{B,2} + O(\delta^3),$$

in the sense that

$$\|\mathcal{W}_B(\delta) - \mathcal{W}_{B,0} - \delta \mathcal{W}_{B,1} - \delta^2 \mathcal{W}_{B,2}\| \leq C \delta^3,$$

for a constant  $C$  independent of  $\delta$ . Here  $\|A\| = \sup_{i,j} \|A_{i,j}\|_{H_T^{-\frac{1}{2}}(\text{div}, \partial B)}$  for any operator-valued matrix  $A$  with entries  $A_{i,j}$ .

We are now interested in finding  $\mathcal{W}_B^{-1}(\delta)$ . For this purpose, we first consider solving the problem

$$(\lambda Id - \mathcal{M}_B) [\psi] = \varphi \quad (3.9)$$

for  $(\psi, \varphi) \in (H_T^{-\frac{1}{2}}(\text{div}, \partial B))^2$  and  $\lambda \notin \sigma(\mathcal{M}_B)$ , where  $\sigma(\mathcal{M}_B)$  is the spectrum of  $\mathcal{M}_B$ .

Using the Helmholtz decomposition of  $H_T^{-\frac{1}{2}}(\text{div}, \partial B)$  in Lemma 2.2, we can reduce (3.9) to an equivalent system of equations involving some well known operators.

**Definition 1.** For  $u \in H_T^{-\frac{1}{2}}(\text{div}, \partial B)$ , we denote by  $u^{(1)}$  and  $u^{(2)}$  any two functions in  $H_0^{\frac{3}{2}}(\partial B)$  and  $H^{\frac{1}{2}}(\partial B)$ , respectively, such that

$$u = \nabla_{\partial B} u^{(1)} + \vec{\text{curl}}_{\partial B} u^{(2)}.$$

Note that  $u^{(1)}$  is uniquely defined and  $u^{(2)}$  is defined up to the sum of a constant function.

**Lemma 3.3.** Assume  $\lambda \neq \frac{1}{2}$ , then problem (3.9) is equivalent to

$$(\lambda Id - \widetilde{\mathcal{M}}_B) \begin{pmatrix} \psi^{(1)} \\ \psi^{(2)} \end{pmatrix} = \begin{pmatrix} \varphi^{(1)} \\ \varphi^{(2)} \end{pmatrix}, \quad (3.10)$$

where  $(\varphi^{(1)}, \varphi^{(2)}) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$  and

$$\widetilde{\mathcal{M}}_B = \begin{pmatrix} -\Delta_{\partial B}^{-1} \mathcal{K}_B^* \Delta_{\partial B} & 0 \\ \mathcal{R}_B & \mathcal{K}_B \end{pmatrix}.$$

*Proof.* Let  $(\psi^{(1)}, \psi^{(2)}) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$  be a solution (if there is any) to (3.10) where  $(\varphi^{(1)}, \varphi^{(2)}) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$  satisfies

$$\varphi = \nabla_{\partial B} \varphi^{(1)} + \vec{\text{curl}}_{\partial B} \varphi^{(2)}.$$

We have

$$(\lambda Id + \Delta_{\partial B}^{-1} \mathcal{K}_B^* \Delta_{\partial B})[\psi^{(1)}] = \varphi^{(1)} \quad (3.11)$$

$$\lambda \psi^{(2)} - \mathcal{R}_B[\psi^{(1)}] - \mathcal{K}_B[\psi^{(2)}] = \varphi^{(2)}. \quad (3.12)$$

Taking  $\nabla_{\partial B}$  in (3.11),  $\vec{\text{curl}}_{\partial B}$  in (3.12), adding up and using the identities of Lemma 2.4 yields

$$(\lambda Id - \mathcal{M}_B) [\nabla_{\partial B} \psi^{(1)} + \vec{\text{curl}}_{\partial B} \psi^{(2)}] = \nabla_{\partial B} \varphi^{(1)} + \vec{\text{curl}}_{\partial B} \varphi^{(2)}.$$

Therefore

$$\psi = \nabla_{\partial B} \psi^{(1)} + \vec{\text{curl}}_{\partial B} \psi^{(2)},$$

is a solution of (3.9).

Conversely, let  $\psi$  be the solution to (3.9). There exist  $(\psi^{(1)}, \psi^{(2)}) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$  and  $(\varphi^{(1)}, \varphi^{(2)}) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$  such that

$$\begin{aligned} \psi &= \nabla_{\partial B} \psi^{(1)} + \vec{\text{curl}}_{\partial B} \psi^{(2)}, \\ \varphi &= \nabla_{\partial B} \varphi^{(1)} + \vec{\text{curl}}_{\partial B} \varphi^{(2)}. \end{aligned}$$

and we have

$$(\lambda Id - \mathcal{M}_B) [\nabla_{\partial B} \psi^{(1)} + \vec{\text{curl}}_{\partial B} \psi^{(2)}] = \nabla_{\partial B} \varphi^{(1)} + \vec{\text{curl}}_{\partial B} \varphi^{(2)}. \quad (3.13)$$

Taking  $\nabla_{\partial B} \cdot$  in the above equation and using the identities of Lemma 2.4 yields

$$\Delta_{\partial B} (\lambda Id + \Delta_{\partial B}^{-1} \mathcal{K}_B^* \Delta_{\partial B}) [\psi^{(1)}] = \Delta_{\partial B} \varphi^{(1)}.$$

Since  $(\psi^{(1)}, \varphi^{(1)}) \in (H_0^{\frac{3}{2}}(\partial B))^2$  we get

$$(\lambda Id + \Delta_{\partial B}^{-1} \mathcal{K}_B^* \Delta_{\partial B}) [\psi^{(1)}] = \varphi^{(1)}.$$

Taking  $\text{curl}_{\partial B}$  in (3.13) and using the identities of Lemma 2.4 yields

$$\Delta_{\partial B} (\lambda \psi^{(2)} - \mathcal{R}_B[\psi^{(1)}] - \mathcal{K}_B[\psi^{(2)}]) = \Delta_{\partial B} \varphi^{(2)}.$$

Therefore there exists a constant  $c$  such that

$$\lambda\psi^{(2)} - \mathcal{R}_B[\psi^{(1)}] - \mathcal{K}_B[\psi^{(2)}] = \varphi^{(2)} + c\chi(\partial B).$$

Since  $\mathcal{K}_B(\chi(\partial B)) = \frac{1}{2}\chi(\partial B)$  we have

$$\lambda\left(\psi^{(2)} - \frac{c}{\lambda - 1/2}\right) - \mathcal{R}_B[\psi^{(1)}] - \mathcal{K}_B\left[\psi^{(2)} - \frac{c}{\lambda - 1/2}\right] = \varphi^{(2)}.$$

Hence,  $\left(\psi^{(1)}, \psi^{(2)} - \frac{c}{\lambda - 1/2}\right) \in H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$  is a solution to (3.10)  $\square$

Let us now analyze the spectral properties of  $\widetilde{\mathcal{M}}_B$  in

$$H(\partial B) := H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B) \quad (3.14)$$

equipped with the inner product

$$(u, v)_{H(\partial B)} = (\Delta_{\partial B} u^{(1)}, \Delta_{\partial B} v^{(1)})_{\mathcal{H}^*} + (u^{(2)}, v^{(2)})_{\mathcal{H}},$$

which is equivalent to  $H_0^{\frac{3}{2}}(\partial B) \times H^{\frac{1}{2}}(\partial B)$ .

By abuse of notation we call  $u^{(1)}$  and  $u^{(2)}$  the first and second components of any  $u \in H(\partial B)$ . We will assume for simplicity the following condition.

**Condition 1.** *The eigenvalues of  $\mathcal{K}_B^*$  are simple.*

Recall that  $\mathcal{K}_B^*$  and  $\mathcal{K}_B$  are compact and self-adjoint in  $\mathcal{H}^*(\partial B)$  and  $\mathcal{H}(\partial B)$ , respectively. Since  $\mathcal{K}_B$  is the  $(\cdot, \cdot)_{-\frac{1}{2}, \frac{1}{2}}$  adjoint of  $\mathcal{K}_B^*$ , we have  $\sigma(\mathcal{K}_B) = \sigma(\mathcal{K}_B^*)$ , where  $\sigma(\mathcal{K}_B)$  (resp.  $\sigma(\mathcal{K}_B^*)$ ) is the (discrete) spectrum of  $\mathcal{K}_B$  (resp.  $\mathcal{K}_B^*$ ). Define

$$\begin{aligned} \sigma_1 &= \sigma(-\mathcal{K}_B^*) \setminus \left(\sigma(\mathcal{K}_B) \cup \left\{-\frac{1}{2}\right\}\right), \\ \sigma_2 &= \sigma(\mathcal{K}_B) \setminus \sigma(-\mathcal{K}_B^*), \\ \sigma_3 &= \sigma(-\mathcal{K}_B^*) \cap \sigma(\mathcal{K}_B). \end{aligned} \quad (3.15)$$

Let  $\lambda_{j,1} \in \sigma_1$ ,  $j = 1, 2, \dots$  and let  $\varphi_{j,1}$  be an associated normalized eigenfunction of  $\mathcal{K}_B^*$  as defined in Lemma 2.1. Note that  $\varphi_{j,1} \in H_0^{-\frac{1}{2}}(\partial B)$  for  $j \geq 1$ . Then,

$$\psi_{j,1} = \begin{pmatrix} \Delta_{\partial B}^{-1} \varphi_{j,1} \\ (\lambda_{j,1} Id - \mathcal{K}_B)^{-1} \mathcal{R}_B[\Delta_{\partial B}^{-1} \varphi_{j,1}] \end{pmatrix}$$

satisfies

$$\widetilde{\mathcal{M}}_B[\psi_{j,1}] = \lambda_{j,1} \psi_{j,1}.$$

Let  $\lambda_{j,2} \in \sigma_2$  and let  $\varphi_{j,2}$  be an associated normalized eigenfunction of  $\mathcal{K}_B$ . Then,

$$\psi_{j,2} = \begin{pmatrix} 0 \\ \varphi_{j,2} \end{pmatrix}$$

satisfies

$$\widetilde{\mathcal{M}}_B[\psi_{j,2}] = \lambda_{j,2}\psi_{j,2}.$$

Now, assume that Condition 1 holds. Let  $\lambda_{j,3} \in \sigma_3$ , let  $\varphi_{j,3}^{(1)}$  be the associated normalized eigenfunction of  $\mathcal{K}_B^*$  and let  $\varphi_{j,3}^{(2)}$  be the associated normalized eigenfunction of  $\mathcal{K}_B$ . Then,

$$\psi_{j,3} = \begin{pmatrix} 0 \\ \varphi_{j,3}^{(2)} \end{pmatrix}$$

satisfies

$$\widetilde{\mathcal{M}}_B[\psi_{j,3}] = \lambda_{j,3}\psi_{j,3},$$

and  $\lambda_{j,3}$  has a first-order generalized eigenfunction given by

$$\psi_{j,3,g} = \begin{pmatrix} c\Delta_{\partial B}^{-1}\varphi_{j,3}^{(1)} \\ (\lambda_{j,3}Id - \mathcal{K}_B)^{-1}\mathcal{P}_{\text{span}\{\varphi_{j,3}^{(2)}\}^\perp}\mathcal{R}_B[c\Delta_{\partial B}^{-1}\varphi_{j,3}^{(1)}] \end{pmatrix} \quad (3.16)$$

for a constant  $c$  such that  $\mathcal{P}_{\text{span}\{\varphi_{j,3}^{(2)}\}}\mathcal{R}_B[c\Delta_{\partial B}^{-1}\varphi_{j,3}^{(1)}] = -\varphi_{j,3}^{(2)}$ . Here,  $\text{span}\{\varphi_{j,3}^{(2)}\}$  is the vector space spanned by  $\varphi_{j,3}^{(2)}$ ,  $\text{span}\{\varphi_{j,3}^{(2)}\}^\perp$  is the orthogonal space to  $\text{span}\{\varphi_{j,3}^{(2)}\}$  in  $\mathcal{H}(\partial B)$  (Lemma 2.1), and  $\mathcal{P}_{\text{span}\{\varphi_{j,3}^{(2)}\}}$  (resp.  $\mathcal{P}_{\text{span}\{\varphi_{j,3}^{(2)}\}^\perp}$ ) is the orthogonal (in  $\mathcal{H}(\partial B)$ ) projection on  $\text{span}\{\varphi_{j,3}^{(2)}\}$  (resp.  $\text{span}\{\varphi_{j,3}^{(2)}\}^\perp$ ).

We remark that the function  $\psi_{j,3,g}$  is determined by the following equation

$$\widetilde{\mathcal{M}}_B[\psi_{j,3,g}] = \lambda_{j,3}\psi_{j,3,g} + \psi_{j,3}.$$

Consequently, the following result holds.

**Proposition 3.1.** *The spectrum  $\sigma(\widetilde{\mathcal{M}}_B) = \sigma_1 \cup \sigma_2 \cup \sigma_3 = \sigma(-\mathcal{K}_B^*) \cup \sigma(\mathcal{K}_B^*) \setminus \{-\frac{1}{2}\}$  in  $H(\partial B)$ .*

*Moreover, under Condition 1,  $\widetilde{\mathcal{M}}_B$  has eigenfunctions  $\psi_{j,i}$  associated to the eigenvalues  $\lambda_{j,i} \in \sigma_i$  for  $j = 1, 2, \dots$  and  $i = 1, 2, 3$ , and generalized eigenfunctions of order one  $\psi_{j,3,g}$  associated to  $\lambda_{j,3} \in \sigma_3$ , all of which form a non-orthogonal basis of  $H(\partial B)$  (defined by (3.14)).*

*Proof.* It is clear that  $\lambda - \widetilde{\mathcal{M}}_B$  is bijective if and only if  $\lambda \notin \sigma(-\mathcal{K}_B^*) \cup \sigma(\mathcal{K}_B^*) \setminus \{-\frac{1}{2}\}$ .

It is only left to show that  $\psi_{j,1}, \psi_{j,2}, \psi_{j,3}, \psi_{j,3,g}$ ,  $j = 1, 2, \dots$  form a non-orthogonal basis of  $H(\partial B)$ .

Indeed, let

$$\psi = \begin{pmatrix} \psi^{(1)} \\ \psi^{(2)} \end{pmatrix} \in H(\partial B).$$

Since  $\psi_{j,1}^{(1)} \cup \psi_{j,3,g}^{(1)}$ ,  $j = 1, 2, \dots$  form an orthogonal basis of  $\mathcal{H}_0^*(\partial B)$ , which is equivalent to  $H_0^{-\frac{1}{2}}(\partial B)$ , there exist  $\alpha_\kappa, \kappa \in I_1 := \{(j, 1) \cup (j, 3, g) : j = 1, 2, \dots\}$  such that

$$\psi^{(1)} = \sum_{\kappa \in I_1} \alpha_\kappa \Delta_{\partial B}^{-1} \psi_\kappa^{(1)},$$

and

$$\sum_{\kappa \in I_1} |\alpha_\kappa|^2 \leq \infty.$$

It is clear that  $\|\psi_\kappa^{(2)}\|_{\mathcal{L}(H^{\frac{1}{2}}(\partial B))}$  is uniformly bounded with respect to  $\kappa \in I_1$ . Then

$$h := \sum_{\kappa \in I_1} \alpha_\kappa \psi_\kappa^{(2)} \in H^{\frac{1}{2}}(\partial B).$$

Since  $\psi_{j,2}^{(2)} \cup \psi_{j,3}^{(2)}$ ,  $j = 1, 2, \dots$  form an orthogonal basis of  $\mathcal{H}(\partial B)$ , which is equivalent to  $H^{\frac{1}{2}}(\partial B)$ , there exist  $\alpha_\kappa, \kappa \in I_2 := \{(j, 2) \cup (j, 3) : j = 1, 2, \dots\}$  such that

$$\psi^{(2)} - h = \sum_{\kappa \in I_2} \alpha_\kappa \psi_\kappa^{(2)},$$

and

$$\sum_{\kappa \in I_2} |\alpha_\kappa|^2 \leq \infty.$$

Hence, there exist  $\alpha_\kappa, \kappa \in I_1 \cup I_2$  such that

$$\psi = \sum_{\kappa \in I_1 \cup I_2} \alpha_\kappa \psi_\kappa,$$

and

$$\sum_{\kappa \in I_1 \cup I_2} |\alpha_\kappa|^2 \leq \infty.$$

□

To have the compactness of  $\widetilde{\mathcal{M}}_B$  we need the following condition

**Condition 2.**  $\sigma_3$  is finite.

Indeed, if  $\sigma_3$  is not finite we have  $\widetilde{\mathcal{M}}_B(\{\psi_{j,3,g}; j \geq 1\}) = \{\lambda_{j,3} \psi_{j,g,3} + \psi_{j,3}; j \geq 1\}$  whose adherence is not compact. However, if  $\sigma_3$  is finite, using Proposition 3.1 we can approximate

$\widetilde{\mathcal{M}}_B$  by a sequence of finite-rank operators.

Throughout this paper, we assume that Condition 2 holds, even though an analysis can still be done for the case where  $\sigma_3$  is infinite; see section 6.

**Definition 2.** Let  $\mathcal{B}$  be the basis of  $H(\partial B)$  formed by the eigenfunctions and generalized eigenfunctions of  $\widetilde{\mathcal{M}}_B$  as stated in Lemma 3.1. For  $\psi \in H(\partial B)$ , we denote by  $\alpha(\psi, \psi_\kappa)$  the projection of  $\psi$  into  $\psi_\kappa \in \mathcal{B}$  such that

$$\psi = \sum_{\kappa} \alpha(\psi, \psi_\kappa) \psi_\kappa.$$

The following lemma follows from the Fredholm alternative.

**Lemma 3.4.** *Let*

$$\psi = \begin{pmatrix} \psi^{(1)} \\ \psi^{(2)} \end{pmatrix} \in H(\partial B).$$

Then,

$$\alpha(\psi, \psi_\kappa) = \begin{cases} \frac{(\psi, \widetilde{\psi}_\kappa)_{H(\partial B)}}{(\psi_\kappa, \widetilde{\psi}_\kappa)_{H(\partial B)}} & \kappa = (j, i), i = 1, 2, \\ \frac{(\psi, \widetilde{\psi}_{\kappa'})_{H(\partial B)}}{(\psi_\kappa, \widetilde{\psi}_{\kappa'})_{H(\partial B)}} & \kappa = (j, 3, g), \kappa' = (j, 3), \\ \frac{(\psi, \widetilde{\psi}_{\kappa_g})_{H(\partial B)} - \alpha(\psi, \psi_{\kappa_g})(\psi_{\kappa_g}, \widetilde{\psi}_{\kappa_g})_{H(\partial B)}}{(\psi_\kappa, \widetilde{\psi}_{\kappa_g})_{H(\partial B)}} & \kappa = (j, 3), \kappa_g = (j, 3, g), \end{cases}$$

where  $\widetilde{\psi}_\kappa \in \text{Ker}(\bar{\lambda}_\kappa - \mathcal{M}_B^*)$  for  $\kappa = (j, i)$ ,  $i = 1, 2, 3$ ;  $\widetilde{\psi}_\kappa \in \text{Ker}(\bar{\lambda}_\kappa - \mathcal{M}_B^*)^2$  for  $\kappa = (j, 3, g)$  and  $\mathcal{M}_B^*$  is the  $H(\partial B)$ -adjoint of  $\mathcal{M}_B$ .

The following remark is in order.

**Remark 3.1.** Note that, since  $\varphi_{j,1}$  and  $\varphi_{j,3}^{(1)}$  form an orthogonal basis of  $\mathcal{H}_0^*(\partial B)$ , equivalent to  $H_0^{-\frac{1}{2}}(\partial B)$ , we also have

$$\alpha(\psi, \psi_\kappa) = \begin{cases} (\Delta_{\partial B} \psi^{(1)}, \varphi_{j,1})_{\mathcal{H}^*} & \kappa = (j, 1), \\ \frac{1}{c} (\Delta_{\partial B} \psi^{(1)}, \varphi_{j,3}^{(1)})_{\mathcal{H}^*} & \kappa = (j, 3, g), \end{cases}$$

where  $c$  is defined in (3.16).

**Remark 3.2.** For  $i = 1, 2, 3$ , and  $j = 1, 2, \dots$ ,

$$\begin{aligned} (\lambda Id - \widetilde{\mathcal{M}}_B)^{-1}[\psi_{j,i}] &= \frac{\psi_{j,i}}{\lambda - \lambda_{j,i}}, \\ (\lambda Id - \widetilde{\mathcal{M}}_B)^{-1}[\psi_{j,3,g}] &= \frac{\psi_{j,3,g}}{\lambda - \lambda_{j,3}} + \frac{\psi_{j,3}}{(\lambda - \lambda_{j,3})^2}. \end{aligned}$$

Now we turn to the original equation (3.4). The following result holds.

**Lemma 3.5.** *The system of equations (3.4) is equivalent to*

$$W_B(\delta) \begin{pmatrix} \eta(\psi)^{(1)} \\ \eta(\psi)^{(2)} \\ \omega\eta(\phi)^{(1)} \\ \omega\eta(\phi)^{(2)} \end{pmatrix} = \left( \begin{array}{c} \frac{\eta(\nu \times E^i)^{(1)}}{\mu_m - \mu_c} \\ \frac{\eta(\nu \times E^i)^{(2)}}{\mu_m - \mu_c} \\ \frac{\eta(i\nu \times H^i)^{(1)}}{\varepsilon_m - \varepsilon_c} \\ \frac{\eta(i\nu \times H^i)^{(2)}}{\varepsilon_m - \varepsilon_c} \end{array} \right) \Big|_{\partial B}, \quad (3.17)$$

where

$$W_B(\delta) = W_{B,0} + \delta W_{B,1} + \delta^2 W_{B,2} + O(\delta^3)$$

with

$$\begin{aligned} W_{B,0} &= \begin{pmatrix} \lambda_\mu Id - \widetilde{\mathcal{M}}_B & O \\ O & \lambda_\varepsilon Id - \widetilde{\mathcal{M}}_B \end{pmatrix}, \\ W_{B,1} &= \begin{pmatrix} O & \frac{1}{\mu_m - \mu_c} \widetilde{\mathcal{L}}_{B,1} \\ \frac{1}{\varepsilon_m - \varepsilon_c} \widetilde{\mathcal{L}}_{B,1} & O \end{pmatrix}, \\ W_{B,2} &= \begin{pmatrix} \frac{1}{\mu_m - \mu_c} \widetilde{\mathcal{M}}_{B,2}^\mu & \frac{1}{\mu_m - \mu_c} \widetilde{\mathcal{L}}_{B,2} \\ \frac{1}{\varepsilon_m - \varepsilon_c} \widetilde{\mathcal{L}}_{B,2} & \frac{1}{\varepsilon_m - \varepsilon_c} \widetilde{\mathcal{M}}_{B,2}^\varepsilon \end{pmatrix} \end{aligned}$$

and

$$\begin{aligned} \widetilde{\mathcal{M}}_B &= \begin{pmatrix} -\Delta_{\partial B}^{-1} \mathcal{K}_B^* \Delta_{\partial B} & 0 \\ \mathcal{R}_B & \mathcal{K}_B \end{pmatrix}, \\ \widetilde{\mathcal{M}}_{B,2}^\mu &= \begin{pmatrix} \Delta_{\partial B}^{-1} \nabla_{\partial B} \cdot (\mu_m \mathcal{M}_{B,2}^{k_m} - \mu_c \mathcal{M}_{B,2}^{k_c}) \nabla_{\partial B} & \Delta_{\partial B}^{-1} \nabla_{\partial B} \cdot (\mu_m \mathcal{M}_{B,2}^{k_m} - \mu_c \mathcal{M}_{B,2}^{k_c}) \vec{\text{curl}}_{\partial B} \\ -\Delta_{\partial B}^{-1} \text{curl}_{\partial B} (\mu_m \mathcal{M}_{B,2}^{k_m} - \mu_c \mathcal{M}_{B,2}^{k_c}) \nabla_{\partial B} & -\Delta_{\partial B}^{-1} \text{curl}_{\partial B} (\mu_m \mathcal{M}_{B,2}^{k_m} - \mu_c \mathcal{M}_{B,2}^{k_c}) \vec{\text{curl}}_{\partial B} \end{pmatrix}, \\ \widetilde{\mathcal{M}}_{B,2}^\varepsilon &= \begin{pmatrix} \Delta_{\partial B}^{-1} \nabla_{\partial B} \cdot (\varepsilon_m \mathcal{M}_{B,2}^{k_m} - \varepsilon_c \mathcal{M}_{B,2}^{k_c}) \nabla_{\partial B} & \Delta_{\partial B}^{-1} \nabla_{\partial B} \cdot (\varepsilon_m \mathcal{M}_{B,2}^{k_m} - \varepsilon_c \mathcal{M}_{B,2}^{k_c}) \vec{\text{curl}}_{\partial B} \\ -\Delta_{\partial B}^{-1} \text{curl}_{\partial B} (\varepsilon_m \mathcal{M}_{B,2}^{k_m} - \varepsilon_c \mathcal{M}_{B,2}^{k_c}) \nabla_{\partial B} & -\Delta_{\partial B}^{-1} \text{curl}_{\partial B} (\varepsilon_m \mathcal{M}_{B,2}^{k_m} - \varepsilon_c \mathcal{M}_{B,2}^{k_c}) \vec{\text{curl}}_{\partial B} \end{pmatrix}, \\ \widetilde{\mathcal{L}}_{B,s} &= \begin{pmatrix} \Delta_{\partial B}^{-1} \nabla_{\partial B} \cdot \mathcal{L}_{B,s} \nabla_{\partial B} & \Delta_{\partial B}^{-1} \nabla_{\partial B} \cdot \mathcal{L}_{B,s} \vec{\text{curl}}_{\partial B} \\ -\Delta_{\partial B}^{-1} \text{curl}_{\partial B} \mathcal{L}_{B,s} \nabla_{\partial B} & -\Delta_{\partial B}^{-1} \text{curl}_{\partial B} \mathcal{L}_{B,s} \vec{\text{curl}}_{\partial B} \end{pmatrix}, \end{aligned}$$

for  $s = 1, 2$ .

Moreover, the eigenfunctions of  $W_{B,0}$  in  $H(\partial B)^2$  are given by

$$\begin{aligned} \Psi_{1,j,i} &= \begin{pmatrix} \psi_{j,i} \\ O \end{pmatrix} \quad j = 0, 1, 2, \dots; i = 1, 2, 3, \\ \Psi_{2,j,i} &= \begin{pmatrix} O \\ \psi_{j,i} \end{pmatrix} \quad j = 0, 1, 2, \dots; i = 1, 2, 3, \end{aligned}$$

associated to the eigenvalues  $\lambda_\mu - \lambda_{j,i}$  and  $\lambda_\varepsilon - \lambda_{j,i}$ , respectively, and generalized eigenfunctions of order one

$$\begin{aligned}\Psi_{1,j,3,g} &= \begin{pmatrix} \psi_{j,3,g} \\ O \end{pmatrix}, \\ \Psi_{2,j,3,g} &= \begin{pmatrix} O \\ \psi_{j,3,g} \end{pmatrix},\end{aligned}$$

associated to eigenvalues  $\lambda_\mu - \lambda_{j,3}$  and  $\lambda_\varepsilon - \lambda_{j,3}$ , respectively, all of which form a non-orthogonal basis of  $H(\partial B)^2$ .

*Proof.* The proof follows directly from Lemmas 3.3 and 3.1.  $\square$

We regard the operator  $W_B(\delta)$  as a perturbation of the operator  $W_{B,0}$  for small  $\delta$ . Using perturbation theory, we can derive the perturbed eigenvalues and their associated eigenfunctions in  $H(\partial B)^2$ .

We denote by  $\Gamma = \{(k, j, i) : k = 1, 2; j = 1, 2, \dots; i = 1, 2, 3\}$  the set of indices for the eigenfunctions of  $W_{B,0}$  and by  $\Gamma_g = \{(k, j, 3, g) : k = 1, 2; j = 1, 2, \dots\}$  the set of indices for the generalized eigenfunctions. We denote by  $\gamma_g$  the generalized eigenfunction index corresponding to eigenfunction index  $\gamma$  and vice-versa. We also denote by

$$\tau_\gamma = \begin{cases} \lambda_\mu - \lambda_{j,i} & k = 1, \\ \lambda_\varepsilon - \lambda_{j,i} & k = 2. \end{cases} \quad (3.18)$$

**Condition 3.**  $\lambda_\mu \neq \lambda_\varepsilon$ .

In the following we will only consider  $\gamma \in \Gamma$  with which there is no generalized eigenfunction index associated. In other words, we only consider  $\gamma = (k, i, j) \in \Gamma$  such that  $\lambda_{j,i} \in \sigma_1 \cup \sigma_2$  (see (3.15) for the definitions). We call this subset  $\Gamma_{\text{sim}}$ .

Note that Conditions 1 and 3 imply that the eigenvalues of  $W_{B,0}$  indexed by  $\gamma \in \Gamma_{\text{sim}}$  are simple. As  $\delta$  goes to zero, the perturbed eigenvalues and eigenfunctions indexed by  $\gamma \in \Gamma_{\text{sim}}$  have the following asymptotic expansions:

$$\begin{aligned}\tau_\gamma(\delta) &= \tau_\gamma + \delta\tau_{\gamma,1} + \delta^2\tau_{\gamma,2} + O(\delta^3), \\ \Psi_\gamma(\delta) &= \Psi_\gamma + \delta\Psi_{\gamma,1} + O(\delta^2),\end{aligned} \quad (3.19)$$

where

$$\begin{aligned}\tau_{\gamma,1} &= \frac{(W_{B,1}\Psi_\gamma, \tilde{\Psi}_\gamma)_{H(\partial B)^2}}{(\Psi_\gamma, \tilde{\Psi}_\gamma)_{H(\partial B)^2}} = 0, \\ \tau_{\gamma,2} &= \frac{(W_{B,2}\Psi_\gamma, \tilde{\Psi}_\gamma)_{H(\partial B)^2} - (W_{B,1}\Psi_{\gamma,1}, \tilde{\Psi}_\gamma)_{H(\partial B)^2}}{(\Psi_\gamma, \tilde{\Psi}_\gamma)_{H(\partial B)^2}}, \\ (\tau_\gamma - W_{B,0})\Psi_{\gamma,1} &= -W_{B,1}\Psi_\gamma.\end{aligned} \quad (3.20)$$

Here,  $\tilde{\Psi}_{\gamma'} \in \text{Ker}(\bar{\tau}_{\gamma'} - W_{B,0}^*)$  and  $W_{B,0}^*$  is the  $H(\partial B)^2$  adjoint of  $W_{B,0}$ .



Using Lemma 3.4 and Remark 3.2 we can solve  $\Psi_{\gamma,1}$ . Indeed,

$$\begin{aligned} \Psi_{\gamma,1} = \sum_{\substack{\gamma' \in \Gamma \\ \gamma' \neq \gamma}} \frac{\alpha(-W_{B,1}\Psi_\gamma, \Psi_{\gamma'})\Psi_{\gamma'}}{\tau_\gamma - \tau_{\gamma'}} + \sum_{\substack{\gamma'_g \in \Gamma_g \\ \gamma'_g \neq \gamma}} \alpha(-W_{B,1}\Psi_\gamma, \Psi_{\gamma'_g}) \left( \frac{\Psi_{\gamma'_g}}{\tau_\gamma - \tau_{\gamma'_g}} + \frac{\Psi_{\gamma'}}{(\tau_\gamma - \tau_{\gamma'})^2} \right) \\ + \alpha(-W_{B,1}\Psi_\gamma, \Psi_\gamma)\Psi_\gamma. \end{aligned}$$

By abuse of notation,

$$\alpha(x, \Psi_\gamma) = \begin{cases} \alpha(x_1, \psi_\kappa) & \gamma = (1, j, i), \kappa = (j, i), \\ \alpha(x_2, \psi_\kappa) & \gamma = (2, j, i), \kappa = (j, i), \end{cases} \quad (3.21)$$

for

$$x = \begin{pmatrix} x_1 \\ x_2 \end{pmatrix} \in H(\partial B)^2,$$

and  $\alpha$  introduced in Definition 2.

Consider now the degenerate case  $\gamma \in \Gamma \setminus \Gamma_{\text{sim}} =: \Gamma_{\text{deg}} = \{\gamma = (k, i, j) \in \Gamma \text{ s.t. } \lambda_{j,i} \in \sigma_3\}$ . It is clear that, for  $\gamma \in \Gamma_{\text{deg}}$ , the algebraic multiplicity of the eigenvalue  $\tau_\gamma$  is 2 while the geometric multiplicity is 1.

In this case every eigenvalue  $\tau_\gamma$  and associated eigenfunction  $\Psi_\gamma$  will split into two branches, as  $\delta$  goes to zero, represented by a convergent Puiseux series as [13]:

$$\begin{aligned} \tau_{\gamma,h}(\delta) &= \tau_\gamma + (-1)^h \delta^{1/2} \tau_{\gamma,1} + (-1)^{2h} \delta^{2/2} \tau_{\gamma,2} + O(\delta^{3/2}), \quad h = 0, 1, \\ \Psi_{\gamma,h}(\delta) &= \Psi_\gamma + (-1)^h \delta^{1/2} \Psi_{\gamma,1} + (-1)^{2h} \delta^{2/2} \Psi_{\gamma,2} + O(\delta^{3/2}), \quad h = 0, 1, \end{aligned} \quad (3.22)$$

where  $\tau_{\gamma,j}$  and  $\Psi_{\gamma,j}$  can be recovered by recurrence formulas. For simplicity we refer to [33] for more details.

### 3.1 First-order correction to plasmonic resonances and field behavior at the plasmonic resonances

Recall that the electric and magnetic parameters,  $\varepsilon_c$  and  $\mu_c$ , depend on the frequency of the incident field,  $\omega$ , following the Drude model [6]. Therefore, the eigenvalues of the operator  $W_{B,0}$  and perturbation in the eigenvalues depend on the frequency as well, that is

$$\begin{aligned} \tau_\gamma(\delta, \omega) &= \tau_\gamma(\omega) + \delta^2 \tau_{\gamma,2}(\omega) + O(\delta^3) \quad \gamma \in \Gamma_{\text{sim}}, \\ \tau_{\gamma,h}(\delta, \omega) &= \tau_\gamma + \delta^{1/2} (-1)^h \tau_{\gamma,1}(\omega) + \delta^{2/2} (-1)^{2h} \tau_{\gamma,2}(\omega) + O(\delta^{3/2}), \quad \gamma \in \Gamma_{\text{deg}}, \quad h = 0, 1. \end{aligned}$$

In the sequel, we will omit frequency dependence to simplify the notation. However, we will keep in mind that all these quantities are frequency dependent.

We first recall different notions of plasmonic resonance [12].

**Definition 3.** (i) We say that  $\omega$  is a plasmonic resonance if  $|\tau_\gamma(\delta)| \ll 1$  and is locally minimized for some  $\gamma \in \Gamma_{\text{sim}}$  or  $|\tau_{\gamma,h}(\delta)| \ll 1$  and is locally minimized for some  $\gamma \in \Gamma_{\text{deg}}$ ,  $h = 0, 1$ .

- (ii) We say that  $\omega$  is a quasi-static plasmonic resonance if  $|\tau_\gamma| \ll 1$  and is locally minimized for some  $\gamma \in \Gamma$ . Here,  $\tau_\gamma$  is defined by (3.18).
- (iii) We say that  $\omega$  is a first-order corrected quasi-static plasmonic resonance if  $|\tau_\gamma + \delta^2 \tau_{\gamma,2}| \ll 1$  and is locally minimized for some  $\gamma \in \Gamma_{\text{sim}}$  or  $|\tau_\gamma + \delta^{1/2}(-1)^h \tau_{\gamma,1}| \ll 1$  and is locally minimized for some  $\gamma \in \Gamma_{\text{deg}}$ ,  $h = 0, 1$ . Here, the correction terms  $\tau_{\gamma,2}$  and  $\tau_{\gamma,1}$  are defined by (3.20) and (3.22).

Note that quasi-static resonance is size independent and is therefore a zero-order approximation of the plasmonic resonance in terms of the particle size while the first-order corrected quasi-static plasmonic resonance depends on the size of the nanoparticle.

We are interested in solving equation (3.17)

$$W_B(\delta)\Psi = f,$$

where

$$\Psi = \begin{pmatrix} \eta(\psi)^{(1)} \\ \eta(\psi)^{(2)} \\ \omega\eta(\phi)^{(1)} \\ \omega\eta(\phi)^{(2)} \end{pmatrix}, f = \begin{pmatrix} \frac{\eta(\nu \times E^i)^{(1)}}{\mu_m - \mu_c} \\ \frac{\eta(\nu \times E^i)^{(2)}}{\mu_m - \mu_c} \\ \frac{\eta(i\nu \times H^i)^{(1)}}{\varepsilon_m - \varepsilon_c} \\ \frac{\eta(i\nu \times H^i)^{(2)}}{\varepsilon_m - \varepsilon_c} \end{pmatrix} \Big|_{\partial B}$$

for  $\omega$  close to the resonance frequencies, i.e., when  $\tau_\gamma(\delta)$  is very small for some  $\gamma$ 's  $\in \Gamma_{\text{sim}}$  or  $\tau_{\gamma,h}(\delta)$  is very small for some  $\gamma$ 's  $\in \Gamma_{\text{deg}}$ ,  $h = 0, 1$ . In this case, the major part of the solution would be the contributions of the excited resonance modes  $\Psi_\gamma(\delta)$  and  $\Psi_{\gamma,h}(\delta)$ .

It is important to remark that problem (3.4) could be ill-posed if either  $\Re(\varepsilon_c) \leq 0$  or  $\Re(\mu_c) \leq 0$  (the imaginary part being very small), and this are precisely the cases for which we will find the resonances described above. In fact, what we do is to solve the problem for the cases  $\Re(\varepsilon_c) > 0$  or  $\Re(\mu_c) > 0$  and then, analytically continue the solution to the general case. The resonances are the values of  $\omega$  for which this analytical continuation "almost" cease to be valid.

We introduce the following definition.

**Definition 4.** We call  $J \subset \Gamma$  index set of resonances if  $\tau_\gamma$ 's are close to zero when  $\gamma \in J$  and are bounded from below when  $\gamma \in J^c$ . More precisely, we choose a threshold number  $\eta_0 > 0$  independent of  $\omega$  such that

$$|\tau_\gamma| \geq \eta_0 > 0 \quad \text{for } \gamma \in J^c.$$

From now on, we shall use  $J$  as our index set of resonances. For simplicity, we assume throughout this paper that the following condition holds.

**Condition 4.** We assume that  $\lambda_\mu \neq 0$ ,  $\lambda_\varepsilon \neq 0$  or equivalently,  $\mu_c \neq -\mu_m$ ,  $\varepsilon_c \neq -\varepsilon_m$ .

It follows that the set  $J$  is finite.

Consider the space  $\mathcal{E}_J = \text{span}\{\Psi_\gamma(\delta), \Psi_{\gamma,h}(\delta); \gamma \in J, h = 0, 1\}$ . Note that, under Condition 4,  $\mathcal{E}_J$  is finite dimensional. Similarly, we define  $\mathcal{E}_{J^c}$  as the spanned by  $\Psi_\gamma(\delta), \Psi_{\gamma,h}(\delta); \gamma \in J^c, h = 0, 1$  and eventually other vectors to complete the base. We have  $H(\partial B)^2 = \mathcal{E}_J \oplus \mathcal{E}_{J^c}$ .

We define  $P_J(\delta)$  and  $P_{J^c}(\delta)$  as the projection into the finite-dimensional space  $\mathcal{E}_J$  and infinite-dimensional space  $\mathcal{E}_{J^c}$ , respectively. It is clear that, for any  $f \in H(\partial B)^2$

$$f = P_J(\delta)[f] + P_{J^c}(\delta)[f].$$

Moreover, we have an explicit representation for  $P_J(\delta)$

$$P_J(\delta)[f] = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \alpha_\delta(f, \Psi_\gamma(\delta)) \Psi_\gamma(\delta) + \sum_{\substack{\gamma \in J \cap \Gamma_{\text{deg}} \\ h=0,1}} \alpha_\delta(f, \Psi_{\gamma,h}(\delta)) \Psi_{\gamma,h}(\delta). \quad (3.23)$$

Here, as in Lemma 3.4,

$$\begin{aligned} \alpha_\delta(f, \Psi_\gamma(\delta)) &= \frac{(f, \tilde{\Psi}_\gamma(\delta))_{H(\partial B)^2}}{(\Psi_\gamma(\delta), \tilde{\Psi}_\gamma(\delta))_{H(\partial B)^2}}, \quad \gamma \in J \cap \Gamma_{\text{sim}}, \\ \alpha_\delta(f, \Psi_{\gamma,h}(\delta)) &= \frac{(f, \tilde{\Psi}_{\gamma,h}(\delta))_{H(\partial B)^2}}{(\Psi_{\gamma,h}(\delta), \tilde{\Psi}_{\gamma,h}(\delta))_{H(\partial B)^2}}, \quad \gamma \in J \cap \Gamma_{\text{deg}}, h = 0, 1. \end{aligned}$$

where  $\tilde{\Psi}_\gamma \in \text{Ker}(\bar{\tau}_{\gamma,h}(\delta) - W_B^*(\delta))$ ,  $\tilde{\Psi}_{\gamma,h} \in \text{Ker}(\bar{\tau}_{\gamma,h}(\delta) - W_B^*(\delta))$  and  $W_B^*(\delta)$  is the  $H(\partial B)^2$ -adjoint of  $W_B(\delta)$ .

We are now ready to solve the equation  $W_B(\delta)\Psi = f$ . In view of Remark 3.2,

$$\Psi = W_B^{-1}(\delta)[f] = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \frac{\alpha_\delta(f, \Psi_\gamma(\delta)) \Psi_\gamma(\delta)}{\tau_\gamma(\delta)} + \sum_{\substack{\gamma \in J \cap \Gamma_{\text{deg}} \\ h=0,1}} \frac{\alpha_\delta(f, \Psi_{\gamma,h}(\delta)) \Psi_{\gamma,h}(\delta)}{\tau_{\gamma,h}(\delta)} + W_B^{-1}(\delta) P_{J^c}(\delta)[f]. \quad (3.24)$$

The following lemma holds.

**Lemma 3.6.** *The norm  $\|W_B^{-1}(\delta) P_{J^c}(\delta)\|_{\mathcal{L}(H(\partial B)^2, H(\partial B)^2)}$  is uniformly bounded in  $\omega$  and  $\delta$ .*

*Proof.* Consider the operator

$$W_B(\delta)|_{J^c} : P_{J^c}(\delta)H(\partial B)^2 \rightarrow P_{J^c}(\delta)H(\partial B)^2.$$

We can show that for every  $\omega$  and  $\delta$ ,  $\text{dist}(\sigma(W_B(\delta)|_{J^c}), 0) \geq \frac{\eta_0}{2}$ , where  $\sigma(W_B(\delta)|_{J^c})$  is the discrete spectrum of  $W_B(\delta)|_{J^c}$ . Here and throughout the paper,  $\text{dist}$  denotes the distance. Then, it follows that

$$\|W_B^{-1}(\delta) P_{J^c}(\delta)[f]\| = \|W_B^{-1}(\delta)|_{J^c} P_{J^c}(\delta)[f]\| \lesssim \frac{1}{\eta_0} \exp\left(\frac{C_1}{\eta_0^2}\right) \|P_{J^c}(\delta)[f]\| \lesssim \frac{1}{\eta_0} \exp\left(\frac{C_1}{\eta_0^2}\right) \|f\|,$$

where the notation  $A \lesssim B$  means that  $A \leq CB$  for some constant  $C$  independent of  $A$  and  $B$ .  $\square$

Finally, we are ready to state our main result in this section.

**Theorem 3.1.** *Let  $\eta$  be defined by (3.5). Under Conditions 1, 2, 3 and 4, the scattered field  $E^s = E - E^i$  due to a single plasmonic particle has the following representation:*

$$E^s = \mu_m \nabla \times \vec{\mathcal{S}}_D^{k_m}[\psi](x) + \nabla \times \nabla \times \vec{\mathcal{S}}_D^{k_m}[\phi](x) \quad x \in \mathbb{R}^3 \setminus \bar{D},$$

where

$$\begin{aligned}\psi &= \eta^{-1}(\nabla_{\partial B}\tilde{\psi}^{(1)} + \vec{\text{curl}}_{\partial B}\tilde{\psi}^{(2)}), \\ \phi &= \frac{1}{\omega}\eta^{-1}(\nabla_{\partial B}\tilde{\phi}^{(1)} + \vec{\text{curl}}_{\partial B}\tilde{\phi}^{(2)}),\end{aligned}$$

$$\Psi = \begin{pmatrix} \tilde{\psi}^{(1)} \\ \tilde{\psi}^{(2)} \\ \tilde{\phi}^{(1)} \\ \tilde{\phi}^{(2)} \end{pmatrix} = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \frac{\alpha(f, \Psi_{\gamma})\Psi_{\gamma} + O(\delta)}{\tau_{\gamma}(\delta)} + \sum_{\gamma \in J \cap \Gamma_{\text{deg}}} \frac{\zeta_1(f)\Psi_{\gamma} + \zeta_2(f)\Psi_{\gamma,1} + O(\delta^{1/2})}{\tau_{\gamma,0}(\delta)\tau_{\gamma,1}(\delta)} + O(1),$$

and

$$\begin{aligned}\zeta_1(f) &= \frac{(f, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2}\tau_{\gamma} - (f, \tilde{\Psi}_{\gamma})_{H(\partial B)^2}(\tau_{\gamma,1} + \tau_{\gamma}\frac{a_2}{a_1})}{a_1}, \\ \zeta_2(f) &= \frac{(f, \tilde{\Psi}_{\gamma})_{H(\partial B)^2}}{a_1}, \\ a_1 &= (\Psi_{\gamma}, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2} + (\Psi_{\gamma,1}, \tilde{\Psi}_{\gamma})_{H(\partial B)^2}, \\ a_2 &= (\Psi_{\gamma}, \tilde{\Psi}_{\gamma,2})_{H(\partial B)^2} + (\Psi_{\gamma,2}, \tilde{\Psi}_{\gamma})_{H(\partial B)^2} + (\Psi_{\gamma,1}, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2}.\end{aligned}$$

*Proof.* Recall that

$$\Psi = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \frac{\alpha_{\delta}(f, \Psi_{\gamma}(\delta))\Psi_{\gamma}(\delta)}{\tau_{\gamma}(\delta)} + \sum_{\substack{\gamma \in J \cap \Gamma_{\text{deg}} \\ h=0,1}} \frac{\alpha_{\delta}(f, \Psi_{\gamma,h}(\delta))\Psi_{\gamma,h}(\delta)}{\tau_{\gamma,h}(\delta)} + W_B^{-1}(\delta)P_{J^c}(\delta)[f].$$

By Lemma 3.6, we have  $W_B^{-1}(\delta)P_{J^c}(\delta)[f] = O(1)$ .

If  $\gamma \in J \cap \Gamma_{\text{sim}}$ , an asymptotic expansion on  $\delta$  yields

$$\alpha_{\delta}(f, \Psi_{\gamma}(\delta))\Psi_{\gamma}(\delta) = \alpha(f, \Psi_{\gamma})\Psi_{\gamma} + O(\delta).$$

If  $\gamma \in J \cap \Gamma_{\text{deg}}$  then  $(\Psi_{\gamma}, \tilde{\Psi}_{\gamma})_{H(\partial B)^2} = 0$ . Therefore, an asymptotic expansion on  $\delta$  yields

$$\begin{aligned}\alpha_{\delta}(f, \Psi_{\gamma,h}(\delta))\Psi_{\gamma,h}(\delta) &= \frac{(-1)^h(f, \tilde{\Psi}_{\gamma})_{H(\partial B)^2}\Psi_{\gamma}}{\delta^{-1/2}a_1} + \\ &\quad \frac{1}{a_1} \left( ((f, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2} - (f, \tilde{\Psi}_{\gamma})_{H(\partial B)^2}\frac{a_2}{a_1})\Psi_{\gamma} + (f, \tilde{\Psi}_{\gamma})_{H(\partial B)^2}\Psi_{\gamma,1} \right) \\ &\quad + O(\delta^{1/2})\end{aligned}$$

with

$$\begin{aligned}a_1 &= (\Psi_{\gamma}, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2} + (\Psi_{\gamma,1}, \tilde{\Psi}_{\gamma})_{H(\partial B)^2}, \\ a_2 &= (\Psi_{\gamma}, \tilde{\Psi}_{\gamma,2})_{H(\partial B)^2} + (\Psi_{\gamma,2}, \tilde{\Psi}_{\gamma})_{H(\partial B)^2} + (\Psi_{\gamma,1}, \tilde{\Psi}_{\gamma,1})_{H(\partial B)^2}.\end{aligned}$$

Since  $\tau_{\gamma,h}(\delta) = \tau_\gamma + \delta^{1/2}(-1)^h \tau_{\gamma,1} + O(\delta)$ , the result follows by adding the terms

$$\frac{\alpha_\delta(f, \Psi_{\gamma,0}(\delta))\Psi_{\gamma,0}(\delta)}{\tau_{\gamma,0}(\delta)} \quad \text{and} \quad \frac{\alpha_\delta(f, \Psi_{\gamma,1}(\delta))\Psi_{\gamma,1}(\delta)}{\tau_{\gamma,1}(\delta)}.$$

The proof is then complete.  $\square$

**Corollary 3.1.** *Assume the same conditions as in Theorem 3.1. Under the additional condition that*

$$\min_{\gamma \in J \cap \Gamma_{\text{sim}}} |\tau_\gamma(\delta)| \gg \delta^3, \quad \min_{\gamma \in J \cap \Gamma_{\text{deg}}} |\tau_\gamma(\delta)| \gg \delta, \quad (3.25)$$

we have

$$\Psi = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \frac{\alpha(f, \Psi_\gamma)\Psi_\gamma + O(\delta)}{\tau_\gamma + \delta^2 \tau_{\gamma,2}} + \sum_{\gamma \in J \cap \Gamma_{\text{deg}}} \frac{\zeta_1(f)\Psi_\gamma + \zeta_2(f)\Psi_{\gamma,1} + O(\delta^{1/2})}{\tau_\gamma^2 - \delta \tau_{\gamma,1}^2} + O(1).$$

**Corollary 3.2.** *Assume the same conditions as in Theorem 3.1. Under the additional condition that*

$$\min_{\gamma \in J \cap \Gamma_{\text{sim}}} |\tau_\gamma(\delta)| \gg \delta^2, \quad \min_{\gamma \in J \cap \Gamma_{\text{deg}}} |\tau_\gamma(\delta)| \gg \delta^{1/2}, \quad (3.26)$$

we have

$$\Psi = \sum_{\gamma \in J \cap \Gamma_{\text{sim}}} \frac{\alpha(f, \Psi_\gamma)\Psi_\gamma + O(\delta)}{\tau_\gamma} + \sum_{\gamma \in J \cap \Gamma_{\text{deg}}} \frac{\alpha(f, \Psi_\gamma)\Psi_\gamma}{\tau_\gamma} + \alpha(f, \Psi_{\gamma,g}) \left( \frac{\Psi_{\gamma,g}}{\tau_\gamma} + \frac{\Psi_\gamma}{\tau_\gamma^2} \right) + O(1).$$

*Proof.* We have

$$\begin{aligned} \lim_{\delta \rightarrow 0} W_B^{-1}(\delta) P_{\text{span}\{\Psi_{\gamma,0}(\delta), \Psi_{\gamma,1}(\delta)\}}[f] &= \lim_{\delta \rightarrow 0} \frac{\alpha_\delta(f, \Psi_{\gamma,0}(\delta))\Psi_{\gamma,0}(\delta)}{\tau_{\gamma,0}(\delta)} + \frac{\alpha_\delta(f, \Psi_{\gamma,1}(\delta))\Psi_{\gamma,1}(\delta)}{\tau_{\gamma,1}(\delta)} \\ &= W_{B,0}^{-1}(\delta) P_{\text{span}\{\Psi_\gamma, \Psi_{\gamma,g}\}}[f] \\ &= \frac{\alpha(f, \Psi_\gamma)\Psi_\gamma}{\tau_\gamma} + \alpha(f, \Psi_{\gamma,g}) \left( \frac{\Psi_{\gamma,g}}{\tau_\gamma} + \frac{\Psi_\gamma}{\tau_\gamma^2} \right), \end{aligned}$$

where  $\gamma \in J \cap \Gamma_{\text{deg}}$ ,  $f \in H(\partial B)^2$  and  $P_{\text{span}E}$  is the projection into the linear space generated by the elements in the set  $E$ .  $\square$

**Remark 3.3.** *Note that for  $\gamma \in J$ ,*

$$\tau_\gamma \approx \min \left\{ \text{dist}(\lambda_\mu, \sigma(\mathcal{K}_B^*) \cup -\sigma(\mathcal{K}_B^*)), \text{dist}(\lambda_\varepsilon, \sigma(\mathcal{K}_B^*) \cup -\sigma(\mathcal{K}_B^*)) \right\}.$$

It is clear, from Remark 3.3, that resonances can occur when exciting the spectrum of  $\mathcal{K}_B^*$  or/and that of  $-\mathcal{K}_B^*$ . We substantiate in the following that only the spectrum of  $\mathcal{K}_B^*$  can be excited to create the plasmonic resonances in the quasi-static regime.

Recall that

$$f = \left( \begin{array}{c} \frac{\eta(\nu \times E^i)^{(1)}}{\mu_m - \mu_c} \\ \frac{\eta(\nu \times E^i)^{(2)}}{\mu_m - \mu_c} \\ \frac{\eta(i\nu \times H^i)^{(1)}}{\varepsilon_m - \varepsilon_c} \\ \frac{\eta(i\nu \times H^i)^{(2)}}{\varepsilon_m - \varepsilon_c} \end{array} \right) \Big|_{\partial B},$$

and therefore,

$$f_1 := \frac{\eta(\nu \times E^i)^{(1)}}{\mu_m - \mu_c} = \frac{\Delta_{\partial B}^{-1} \nabla_{\partial B} \cdot \eta(\nu \times E^i)}{\mu_m - \mu_c}.$$

Now, suppose  $\gamma = (1, j, 1) \in J$  (recall that  $J$  is the index set of resonances). Then  $\tau_\gamma = \lambda_\mu - \lambda_{1,j}$ , where  $\lambda_{1,j} \in \sigma_1 = \sigma(-\mathcal{K}_B^*) \setminus \sigma(\mathcal{K}_B^*)$ . From Remark 3.1,

$$\alpha(f, \Psi_\gamma) = (\Delta_{\partial B} f_1, \varphi_{j,1})_{\mathcal{H}^*} = \alpha(f, \Psi_\gamma) = \frac{1}{\mu_m - \mu_c} (\nabla_{\partial B} \cdot \eta(\nu \times E^i), \varphi_{j,1})_{\mathcal{H}^*},$$

where  $\varphi_{j,1} \in \mathcal{H}_0^*(\partial B)$  is a normalized eigenfunction of  $\mathcal{K}_B^*(\partial B)$ .

A Taylor expansion of  $E^i$  gives, for  $x \in \partial D$ ,

$$E^i(x) = \sum_{\beta \in \mathbb{N}^3} \frac{(x - z)^\beta \partial^\beta E^i(z)}{|\beta|!}.$$

Thus

$$\eta(\nu \times E^i)(\tilde{x}) = \eta(\nu)(\tilde{x}) \times E^i(z) + O(\delta),$$

and

$$\begin{aligned} \nabla_{\partial B} \cdot \eta(\nu \times E^i)(\tilde{x}) &= -\eta(\nu)(\tilde{x}) \cdot \nabla \times E^i(z) + O(\delta) \\ &= O(\delta). \end{aligned}$$

Therefore, the zeroth-order term of the expansion of  $\nabla_{\partial B} \cdot \eta(\nu \times E^i)$  in  $\delta$  is zero. Hence,

$$\alpha(f, \Psi_\gamma) = 0.$$

In the same way, we have

$$\begin{aligned} \alpha(f, \Psi_\gamma) &= 0, \\ \alpha(f, \Psi_{\gamma_g}) &= 0 \end{aligned}$$

for  $\gamma = (2, j, 1) \in J$  and  $\gamma_g$  such that  $\gamma \in J$ .

As a result we see that the spectrum of  $-\mathcal{K}_B^*$  is not excited in the zeroth-order term. However, we note that  $\sigma(-\mathcal{K}_B^*)$  can be excited in higher-order terms.

## 4 The quasi-static limit and the extinction cross-section

### 4.1 The quasi-static limit

In this subsection we recall the quasi-static limit of the electromagnetic field at plasmonic resonances. The formula was first obtained in [6], but it can be derived by pursuing further computations in Corollary 3.2.

We first recall the definition of the polarization tensor

$$M(\lambda, D) = \int_{\partial D} (\lambda Id - \mathcal{K}_D^*)^{-1} [\nu](x) x \, d\sigma(x), \quad (4.1)$$

where  $\lambda \in \mathbb{C} \setminus (-1/2, 1/2)$ . The polarization tensor is a key ingredient of the quasi-static limit, or zeroth-order approximation, of the far-field.

**Theorem 4.1.** *Let  $d_\sigma = \min \left\{ \text{dist}(\lambda_\mu, \sigma(\mathcal{K}_D^*) \cup -\sigma(\mathcal{K}_D^*)), \text{dist}(\lambda_\varepsilon, \sigma(\mathcal{K}_D^*) \cup -\sigma(\mathcal{K}_D^*)) \right\}$ . Then, for  $D = z + \delta B \in \mathbb{R}^3$  of class  $\mathcal{C}^{1,\alpha}$  for  $0 < \alpha < 1$ , the following uniform far-field expansion holds*

$$E^s = -\frac{i\omega\mu_m}{\varepsilon_m} \nabla \times G_d(x, z, k_m) M(\lambda_\mu, D) H^i(z) - \omega^2 \mu_m G_d(x, z, k_m) M(\lambda_\varepsilon, D) E^i(z) + O\left(\frac{\delta^4}{d_\sigma}\right),$$

where

$$G_d(x, z, k_m) = \varepsilon_m (G(x, z, k_m) Id + \frac{1}{k_m^2} D_x^2 G(x, z, k_m))$$

is the Dyadic Green (matrix valued) function for the full Maxwell equations.

### 4.2 The far-field expansion

The following lemma deals with the far-field behavior of the electromagnetic fields. We first recall the representation for the scattering amplitude.

**Lemma 4.1.** *The solution  $(E, H)$  to the system (3.1) has the following far-field expansion:*

$$E^s(x) = -\frac{e^{ik_m|x|}}{4\pi|x|} A_\infty(\hat{x}) + O\left(\frac{1}{|x|^2}\right)$$

as  $|x| \rightarrow +\infty$ , where  $\hat{x} = \frac{x}{|x|}$ ,

$$A_\infty(\hat{x}) = -i\mu_m k_m \hat{x} \times \int_{\partial D} e^{-ik_m \hat{x} \cdot y} \psi(y) d\sigma(y) - k_m^2 \hat{x} \times \hat{x} \times \int_{\partial D} e^{-ik_m \hat{x} \cdot y} \phi(y) d\sigma(y),$$

and

$$H^s(x) = -\frac{e^{ik_m|x|}}{4\pi|x|} \hat{x} \times A_\infty(\hat{x}) + O\left(\frac{1}{|x|^2}\right).$$

The following result is known as the optical cross-section theorem for the scattering of electromagnetic waves [24].

**Theorem 4.2.** Assume that the incident fields are plane waves given by

$$\begin{aligned} E^i(x) &= p e^{ik_m d \cdot x}, \\ H^i(x) &= d \times p e^{ik_m d \cdot x}, \end{aligned}$$

where  $p \in \mathbb{R}^3$  and  $d \in \mathbb{R}^3$  with  $|d| = 1$  are such that  $p \cdot d = 0$ . Then, the extinction cross-section is given by

$$Q^{ext} = \frac{4\pi}{k_m} \Im \left[ \frac{p \cdot A_\infty(d)}{|p|^2} \right],$$

where  $A_\infty$  is the scattering amplitude.

Doing Taylor expansions on the formula of Theorem 4.1 gives the following proposition, which allow us to compute the extinction cross-section in terms of the polarization tensor.

**Proposition 4.1.** Let  $\hat{x} = x/|x|$ . The following far-field asymptotic expansion holds:

$$\begin{aligned} E^s &= -\frac{e^{ik_m|x|}}{4\pi|x|} \left( \omega\mu_m k_m e^{ik_m(d-\hat{x}) \cdot z} (\hat{x} \times Id) M(\lambda_\mu, D)(d \times p) - k_m^2 e^{ik_m(d-\hat{x}) \cdot z} (Id - \hat{x}\hat{x}^t) M(\lambda_\varepsilon, D)p \right) \\ &\quad + O\left(\frac{1}{|x|^2}\right) + O\left(\frac{\delta^4}{d_\sigma}\right). \end{aligned}$$

Then we have, up to an error term of the order  $O(\frac{\delta^4}{d_\sigma})$ ,

$$A_\infty(\hat{x}) = \omega\mu_m k_m e^{ik_m(d-\hat{x}) \cdot z} (\hat{x} \times Id) M(\lambda_\mu, D)(d \times p) - k_m^2 e^{ik_m(d-\hat{x}) \cdot z} (Id - \hat{x}\hat{x}^t) M(\lambda_\varepsilon, D)p.$$

In particular,

$$A_\infty(d) = \omega\mu_m k_m (d \times Id) M(\lambda_\mu, D)(d \times p) - k_m^2 (Id - dd^t) M(\lambda_\varepsilon, D)p,$$

where  $M(\lambda_\mu, D)$  and  $M(\lambda_\varepsilon, D)$  are the polarization tensors associated with  $D$  and  $\lambda = \lambda_\mu$  and  $\lambda = \lambda_\varepsilon$ , respectively.

## 5 Explicit computations for a spherical nanoparticle

### 5.1 Vector spherical harmonics

Let  $\hat{x} = \frac{x}{|x|}$ . For  $m = -n, \dots, n$  and  $n = 1, 2, \dots$ , set  $Y_n^m$  to be the spherical harmonics defined on the unit sphere  $S = \{x \in \mathbb{R}^3, |x| = 1\}$ . For a wave number  $k > 0$ , the function

$$v_{n,m}(k; x) = h_n^{(1)}(k|x|) Y_n^m(\hat{x})$$

satisfies the Helmholtz equation  $\Delta v + k^2 v = 0$  in  $\mathbb{R}^3 \setminus \{0\}$  together with the Sommerfeld radiation condition

$$\lim_{|x| \rightarrow \infty} \left( \frac{\partial v_{n,m}}{\partial |x|}(k; x) - ik v_{n,m}(k; x) \right) = 0.$$



Similarly, let  $\tilde{v}_{n,m}(x)$  be defined by

$$\tilde{v}_{n,m}(x) = j_n(k|x|)Y_n^m(\hat{x}),$$

where  $j_n$  is the spherical Bessel function of the first kind. Then the function  $\tilde{v}_{n,m}$  satisfies the Helmholtz equation in  $\mathbb{R}^3$ .

Next, define the vector spherical harmonics by

$$U_{n,m} = \frac{1}{\sqrt{n(n+1)}} \nabla_S Y_n^m(\hat{x}) \quad \text{and} \quad V_{n,m} = \hat{x} \times U_{n,m}$$

for  $m = -n, \dots, n$  and  $n = 1, 2, \dots$ . Here,  $\hat{x} \in S$  and  $\nabla_S$  denote the surface gradient on the unit sphere  $S$ . The vector spherical harmonics form a complete orthogonal basis for  $L_T^2(S)$ .

Using the vectorial spherical harmonics, we can separate the solutions of Maxwell's equations into multipole solutions; see [41, Section 5.3]. Define the exterior transverse electric multipoles, i.e.,  $E \cdot x = 0$ , as

$$\begin{cases} E_{n,m}^{TE}(x) = -\sqrt{n(n+1)}h_n^{(1)}(k|x|)V_{n,m}(\hat{x}), \\ H_{n,m}^{TE}(x) = -\frac{i}{\omega\mu}\nabla \times \left( -\sqrt{n(n+1)}h_n^{(1)}(k|x|)V_{n,m}(\hat{x}) \right), \end{cases} \quad (5.1)$$

and the exterior transverse magnetic multipoles, i.e.,  $H \cdot x = 0$ , as

$$\begin{cases} E_{n,m}^{TM}(x) = \frac{i}{\omega\epsilon}\nabla \times \left( -\sqrt{n(n+1)}h_n^{(1)}(k|x|)V_{n,m}(\hat{x}) \right), \\ H_{n,m}^{TM}(x) = -\sqrt{n(n+1)}h_n^{(1)}(k|x|)V_{n,m}(\hat{x}). \end{cases} \quad (5.2)$$

The exterior electric and magnetic multipoles satisfy the Sommerfeld radiation condition. In the same manner, one defines the interior multipoles  $(\tilde{E}_{n,m}^{TE}, \tilde{H}_{n,m}^{TE})$  and  $(\tilde{E}_{n,m}^{TM}, \tilde{H}_{n,m}^{TM})$  with  $h_n^{(1)}$  replaced by  $j_n$ , i.e.,

$$\begin{cases} \tilde{E}_{n,m}^{TE}(x) = -\sqrt{n(n+1)}j_n(k|x|)V_{n,m}(\hat{x}), \\ \tilde{H}_{n,m}^{TE}(x) = -\frac{i}{\omega\mu}\nabla \times \tilde{E}_{n,m}^{TE}(x), \end{cases} \quad (5.3)$$

and

$$\begin{cases} \tilde{H}_{n,m}^{TM}(x) = -\sqrt{n(n+1)}j_n(k|x|)V_{n,m}(\hat{x}), \\ \tilde{E}_{n,m}^{TM}(x) = \frac{i}{\omega\epsilon}\nabla \times \tilde{H}_{n,m}^{TM}(x). \end{cases} \quad (5.4)$$

Note that one has

$$\nabla \times E_{n,m}^{TE}(k; x) = \frac{\sqrt{n(n+1)}}{|x|} \mathcal{H}_n(k|x|)U_{n,m}(\hat{x}) + \frac{n(n+1)}{|x|} h_n^{(1)}(k|x|)Y_n^m(\hat{x})\hat{x} \quad (5.5)$$

and

$$\nabla \times \tilde{E}_{n,m}^{TE}(k; x) = \frac{\sqrt{n(n+1)}}{|x|} \mathcal{J}_n(k|x|)U_{n,m}(\hat{x}) + \frac{n(n+1)}{|x|} j_n(k|x|)Y_n^m(\hat{x})\hat{x}, \quad (5.6)$$

where

$$\mathcal{J}_n(t) = j_n(t) + t j_n'(t), \quad \mathcal{H}_n(t) = h_n^{(1)}(t) + t(h_n^{(1)})'(t).$$

For  $|x| > |y|$ , the following addition formula holds:

$$\begin{aligned}
G(x, y, k)I = & - \sum_{n=1}^{\infty} \frac{ik}{n(n+1)} \frac{\epsilon}{\mu} \sum_{m=-n}^n E_{n,m}^{TM}(x) \overline{\widetilde{E}_{n,m}^{TM}(y)}^T \\
& - \sum_{n=1}^{\infty} \frac{ik}{n(n+1)} \sum_{m=-n}^n E_{n,m}^{TE}(x) \overline{\widetilde{E}_{n,m}^{TE}(y)}^T \\
& - \frac{i}{k} \sum_{n=1}^{\infty} \sum_{m=-n}^n \nabla v_{n,m}(x) \overline{\widetilde{\nabla v}_{n,m}(y)}^T.
\end{aligned} \tag{5.7}$$

Alternatively, for  $|x| < |y|$ , we have

$$\begin{aligned}
G(x, y, k)I = & - \sum_{n=1}^{\infty} \frac{ik}{n(n+1)} \frac{\epsilon}{\mu} \sum_{m=-n}^n \overline{\widetilde{E}_{n,m}^{TM}(x)} E_{n,m}^{TM}(y)^T \\
& - \sum_{n=1}^{\infty} \frac{ik}{n(n+1)} \sum_{m=-n}^n \overline{\widetilde{E}_{n,m}^{TE}(x)} E_{n,m}^{TE}(y)^T \\
& - \frac{i}{k} \sum_{n=1}^{\infty} \sum_{m=-n}^n \overline{\widetilde{\nabla v}_{n,m}(x)} \nabla v_{n,m}(y)^T.
\end{aligned} \tag{5.8}$$

## 5.2 Explicit representations of boundary integral operators

Let  $D$  be a sphere of radius  $r > 0$ . We have the following results.

**Lemma 5.1.** *Let  $\partial D = \{|x| = r\}$ . Then, for  $r' > r$ , we have*

$$\nu \times \nabla \times \vec{S}_D^k[U_{n,m}]|_{|x|=r'}^+ = (-ikr)h_n^{(1)}(kr')\mathcal{J}_n(kr)U_{n,m}, \tag{5.9}$$

$$\nu \times \nabla \times \vec{S}_D^k[V_{n,m}]|_{|x|=r'}^+ = ik\frac{r^2}{r'}j_n(kr)\mathcal{H}_n(kr')V_{n,m}, \tag{5.10}$$

$$\nu \times \nabla \times \nabla \times \vec{S}_D^k[U_{n,m}]|_{|x|=r'}^+ = -ik\frac{r}{r'}\mathcal{J}_n(kr)\mathcal{H}_n(kr')V_{n,m}, \tag{5.11}$$

$$\nu \times \nabla \times \nabla \times \vec{S}_D^k[V_{n,m}]|_{|x|=r'}^+ = ik(kr)^2j_n(kr)h_n^{(1)}(kr')U_{n,m}. \tag{5.12}$$

For  $r' < r$ ,

$$\nu \times \nabla \times \vec{S}_D^k[U_{n,m}]|_{|x|=r'}^+ = (-ikr)j_n(kr')\mathcal{H}_n(kr)U_{n,m}, \tag{5.13}$$

$$\nu \times \nabla \times \vec{S}_D^k[V_{n,m}]|_{|x|=r'}^+ = ik\frac{r^2}{r'}\mathcal{J}_n(kr')h_n^{(1)}(kr)V_{n,m}, \tag{5.14}$$

$$\nu \times \nabla \times \nabla \times \vec{S}_D^k[U_{n,m}]|_{|x|=r'}^+ = -ik\frac{r}{r'}\mathcal{J}_n(kr')\mathcal{H}_n(kr)V_{n,m}, \tag{5.15}$$

$$\nu \times \nabla \times \nabla \times \vec{S}_D^k[V_{n,m}]|_{|x|=r'}^+ = ik(kr)^2j_n(kr')h_n^{(1)}(kr)U_{n,m}. \tag{5.16}$$

*Proof.* We only consider (5.9). The other formulas can be proved in a similar way.

From (5.5), (5.6), and the definitions of  $E_{n,m}^{TE}$ ,  $E_{n,m}^{TM}$ ,  $\tilde{E}_{n,m}^{TE}$  and  $\tilde{E}_{n,m}^{TM}$ , we have

$$\begin{aligned}
& \nabla_x \times G(x, y, k) U_{n,m}(\hat{y}) \\
&= - \sum_{n=1}^{\infty} \frac{ik}{n(n+1)} \frac{\epsilon}{\mu} \sum_{m=-n}^n \nabla \times E_{n,m}^{TM}(x) \overline{\tilde{E}_{n,m}^{TM}(y)} \cdot U_{p,q}(\hat{y}) \\
&\quad + \sum_{n=1}^{\infty} \frac{ik}{n(n+1)} \sum_{m=-n}^n \nabla \times E_{n,m}^{TE}(x) \overline{\tilde{E}_{n,m}^{TE}(y)} \cdot U_{p,q}(\hat{y}) \\
&= - \sum_{n=1}^{\infty} \frac{ik}{\sqrt{n(n+1)}} \frac{\epsilon}{\mu} \sum_{m=-n}^n \nabla \times E_{n,m}^{TM}(x) \frac{-i}{\omega \epsilon} \frac{1}{r} \mathcal{J}_n(kr) U_{n,m}(\hat{y}) \cdot U_{p,q}(\hat{y}) \\
&\quad + \sum_{n=1}^{\infty} \frac{ik}{\sqrt{n(n+1)}} \sum_{m=-n}^n \nabla \times E_{n,m}^{TE}(x) (-1) j_n(kr) V_{n,m}(\hat{y}) \cdot U_{p,q}(\hat{y})
\end{aligned}$$

for  $|y| = r$  and  $|x| > |y|$ . Therefore, we get on  $|x| = r$

$$\begin{aligned}
\nabla \times \tilde{S}_D^k[U_{n,m}]|_+ &= \nabla_x \times \int_{|y|=r} G(x, y, k) U_{n,m}(\hat{y}) \\
&= \frac{kr}{\sqrt{n(n+1)}} \frac{1}{\omega \mu} \mathcal{J}_n(kr) (\nabla \times E_{n,m}^{TM}(x))|_{|x|=r}.
\end{aligned} \tag{5.17}$$

Since

$$\nabla \times E_{p,q}^{TM} = \frac{i}{\omega \epsilon} \nabla \times \nabla \times E_{p,q}^{TE} = \frac{i}{\omega \epsilon} k^2 E_{p,q}^{TE},$$

we obtain

$$\begin{aligned}
\hat{x} \times \nabla \times \tilde{S}_D^k[U_{n,m}]|_+ &= \frac{ikr}{\sqrt{n(n+1)}} \mathcal{J}_n(kr) (\hat{x} \times E_{n,m}^{TE}(x))|_{|x|=r} \\
&= (-ikr) h_n^{(1)}(kr) \mathcal{J}_n(kr) U_{n,m} \quad \text{on } |x| = r,
\end{aligned}$$

which completes the proof.  $\square$

Note that

$$\nu \times \nabla \times \tilde{\mathcal{S}}_D^k[\phi]|_{\pm} = (\mp \frac{1}{2} I + \mathcal{M}_D^k)[\phi] \quad \text{on } \partial D,$$

and recall the following identity, which was proved in [46],

$$\nu \times \nabla \times \nabla \times \tilde{\mathcal{S}}_D^k[\phi] = \mathcal{L}_D^k[\phi] \quad \text{on } \partial D.$$

For  $m = -n, \dots, n$  and  $n = 1, 2, 3, \dots$ , let  $H_{n,m}(\partial D)$  be the subspace of  $H(\partial D)$  defined by

$$H_{n,m}(\partial D) = \text{span}\{U_{n,m}, V_{n,m}\}.$$

Let us represent the operators  $\mathcal{M}_D^k$  and  $\mathcal{L}_D^k$  explicitly on the subspace  $H_{m,n}(\partial D)$ . Using  $U_{n,m}, V_{n,m}$  as basis vectors, we obtain the following matrix representations for  $\mathcal{M}_D^k$  and  $\mathcal{L}_D^k$

on the subspace  $H_{n,m}(\partial D)$ :

$$\mathcal{M}_D^k = \begin{pmatrix} \frac{1}{2} - ikrh_n^{(1)}(kr)\mathcal{J}_n(kr) & 0 \\ 0 & \frac{1}{2} + ikrj_n(kr)\mathcal{H}_n(kr) \end{pmatrix}, \quad (5.18)$$

and

$$\mathcal{L}_D^k = \begin{pmatrix} 0 & ik(kr)^2 j_n(kr)h_n^{(1)}(kr) \\ -ik\mathcal{J}_n(kr)\mathcal{H}_n(kr) & 0 \end{pmatrix}. \quad (5.19)$$

### 5.3 Asymptotic behavior of the spectrum of $\mathcal{W}_B(r)$

Now we consider the asymptotic expansions of the operator  $\mathcal{W}_B(r)$  and its spectrum when  $r \ll 1$ .

It is well-known that, as  $t \rightarrow 0$ ,

$$\begin{aligned} j_n(t) &= \frac{t^n}{(2n+1)!!} \left( 1 - \frac{1}{2(2n+3)} t^2 + O(t^4) \right), \\ h_n^{(1)}(t) &= -i((2n-1)!!) t^{-n-1} \left( 1 + \frac{1}{2(2n-1)} t^2 + O(t^4) \right). \end{aligned} \quad (5.20)$$

By making use of these asymptotics of the spherical Bessel functions, we obtain that

$$\begin{aligned} i\mathcal{J}_n(t)h_n^{(1)}(\tilde{t}) &= \frac{n+1}{2n+1} \left( \frac{t}{\tilde{t}} \right)^n \frac{1}{\tilde{t}} + \frac{n+1}{2(2n-1)(2n+1)} \left( \frac{t}{\tilde{t}} \right)^n \tilde{t} - \frac{n+3}{2(2n+1)(2n+3)} \left( \frac{t}{\tilde{t}} \right)^{n+1} t + O(t^3), \\ ij_n(t)\mathcal{H}_n(\tilde{t}) &= \frac{-n}{2n+1} \left( \frac{t}{\tilde{t}} \right)^n \frac{1}{\tilde{t}} + \frac{-n+2}{2(2n-1)(2n+1)} \left( \frac{t}{\tilde{t}} \right)^n \tilde{t} + \frac{n}{2(2n+1)(2n+3)} \left( \frac{t}{\tilde{t}} \right)^{n+1} t + O(t^3), \\ ij_n(t)h_n^{(1)}(\tilde{t}) &= \frac{1}{2n+1} \left( \frac{t}{\tilde{t}} \right)^n \frac{1}{\tilde{t}} + \frac{1}{2(2n-1)(2n+1)} \left( \frac{t}{\tilde{t}} \right)^n \tilde{t} - \frac{1}{2(2n+1)(2n+3)} \left( \frac{t}{\tilde{t}} \right)^{n+1} t + O(t^3), \\ i\mathcal{J}_n(t)\mathcal{H}_n(\tilde{t}) &= \frac{(-n)(n+1)}{2n+1} \left( \frac{t}{\tilde{t}} \right)^n \frac{1}{\tilde{t}} + \frac{(n+1)(-n+2)}{2(2n-1)(2n+1)} \left( \frac{t}{\tilde{t}} \right)^n \tilde{t} + \frac{n(n+3)}{2(2n+1)(2n+3)} \left( \frac{t}{\tilde{t}} \right)^{n+1} t + O(t^3), \end{aligned} \quad (5.21)$$

for small  $t, \tilde{t} \ll 1$  with  $t \approx \tilde{t}$ .

So, we have

$$\mathcal{M}_D^k = \begin{pmatrix} \frac{(-1)}{2(2n+1)} + (kr)^2 r_n & 0 \\ 0 & \frac{1}{2(2n+1)} + (kr)^2 s_n \end{pmatrix} + O(r^4), \quad (5.22)$$

and

$$\mathcal{L}_D^k = \begin{pmatrix} 0 & k^2 r p_n \\ \frac{n(n+1)}{2n+1} \frac{1}{r} + k^2 r q_n & 0 \end{pmatrix} + O(r^3), \quad (5.23)$$

where

$$\begin{aligned}
p_n &= \frac{1}{2n+1}, \\
q_n &= \frac{(n+1)(n-2)}{2(2n-1)(2n+1)} - \frac{n(n+3)}{2(2n+1)(2n+3)}, \\
r_n &= -\frac{n+1}{2(2n-1)(2n+1)} + \frac{(n+3)}{2(2n+1)(2n+3)}, \\
s_n &= -\frac{n-2}{2(2n-1)(2n+1)} + \frac{n}{2(2n+1)(2n+3)}.
\end{aligned} \tag{5.24}$$

Therefore, we can obtain

$$\mathcal{W}_B(r) = \mathcal{W}_{B,0} + r\mathcal{W}_{B,1} + r^2\mathcal{W}_{B,2} + O(r^3),$$

where

$$\mathcal{W}_{B,0} = \begin{pmatrix} \lambda_\mu - \frac{(-1)}{2(2n+1)} & 0 & 0 & 0 \\ 0 & \lambda_\mu - \frac{1}{2(2n+1)} & 0 & 0 \\ 0 & 0 & \lambda_\varepsilon - \frac{(-1)}{2(2n+1)} & 0 \\ 0 & 0 & 0 & \lambda_\varepsilon - \frac{1}{2(2n+1)} \end{pmatrix}, \tag{5.25}$$

$$\mathcal{W}_{B,1} = \begin{pmatrix} 0 & 0 & 0 & \omega C_\mu p_n \\ 0 & 0 & \omega C_\mu q_n & 0 \\ 0 & \omega C_\varepsilon p_n & 0 & 0 \\ \omega C_\varepsilon q_n & 0 & 0 & 0 \end{pmatrix}, \tag{5.26}$$

$$\mathcal{W}_{B,2} = \begin{pmatrix} \omega^2 D_\mu r_n & 0 & 0 & 0 \\ 0 & \omega^2 D_\mu s_n & 0 & 0 \\ 0 & 0 & \omega^2 D_\varepsilon r_n & 0 \\ 0 & 0 & 0 & \omega^2 D_\varepsilon s_n \end{pmatrix}, \tag{5.27}$$

and

$$C_\mu = \frac{\mu_c \varepsilon_c - \mu_m \varepsilon_m}{\mu_m - \mu_c}, \quad C_\varepsilon = \frac{\mu_c \varepsilon_c - \mu_m \varepsilon_m}{\varepsilon_m - \varepsilon_c}, \tag{5.28}$$

$$D_\mu = \frac{\varepsilon_c \mu_c^2 - \varepsilon_m \mu_m^2}{\mu_m - \mu_c}, \quad D_\varepsilon = \frac{\varepsilon_c^2 \mu_c - \varepsilon_m^2 \mu_m}{\varepsilon_m - \varepsilon_c}. \tag{5.29}$$

By applying the standard perturbation theory, the asymptotics of eigenvalues of  $\mathcal{W}_B(r)$  are

obtained as follows: up to an error term of the order  $O(r^3)$ ,

$$\begin{aligned}\lambda_\mu - \frac{(-1)}{2(2n+1)} + (r\omega)^2 \left[ C_\varepsilon C_\mu \frac{p_n q_n}{\lambda_\mu - \lambda_\varepsilon + p_n} + D_\mu r_n \right] + O(r^3), \\ \lambda_\mu - \frac{1}{2(2n+1)} + (r\omega)^2 \left[ C_\varepsilon C_\mu \frac{p_n q_n}{\lambda_\mu - \lambda_\varepsilon - p_n} + D_\mu s_n \right] + O(r^3), \\ \lambda_\varepsilon - \frac{(-1)}{2(2n+1)} + (r\omega)^2 \left[ C_\varepsilon C_\mu \frac{p_n q_n}{\lambda_\varepsilon - \lambda_\mu + p_n} + D_\varepsilon r_n \right] + O(r^3), \\ \lambda_\varepsilon - \frac{1}{2(2n+1)} + (r\omega)^2 \left[ C_\varepsilon C_\mu \frac{p_n q_n}{\lambda_\varepsilon - \lambda_\mu - p_n} + D_\varepsilon s_n \right] + O(r^3),\end{aligned}$$

and the asymptotics of the associated eigenfunction are given by

$$\begin{aligned}[1, 0, 0, 0]^T + r\omega \frac{C_\varepsilon q_n}{\lambda_\mu - \lambda_\varepsilon + p_n} [0, 0, 0, 1]^T + O(r^2), \\ [0, 1, 0, 0]^T + r\omega \frac{C_\varepsilon}{2n+1} \frac{1}{\lambda_\mu - \lambda_\varepsilon - p_n} [0, 0, 1, 0]^T + O(r^2), \\ [0, 0, 1, 0]^T + r\omega \frac{C_\mu q_n}{\lambda_\varepsilon - \lambda_\mu + p_n} [0, 1, 0, 0]^T + O(r^2), \\ [0, 0, 0, 1]^T + r\omega \frac{C_\mu}{2n+1} \frac{1}{\lambda_\varepsilon - \lambda_\mu - p_n} [1, 0, 0, 0]^T + O(r^2).\end{aligned}$$

#### 5.4 Extinction cross-section

In this subsection, we compute the extinction cross-section  $Q^{ext}$ . We need the following lemma.

**Lemma 5.2.** *Let  $D$  be a sphere with radius  $r > 0$  and suppose that  $E^i$  is given by*

$$E^i(x) = \sum_{n=1}^{\infty} \sum_{l=-n}^n \alpha_{nl}^{TE} \tilde{E}_{n,l}^{TE}(x; k_m) + \alpha_{nl}^{TM} \tilde{E}_{n,l}^{TM}(x; k_m),$$

for some coefficients  $\alpha_{nl}^{TE}, \alpha_{nl}^{TM}$ . Then the scattered wave can be represented as follows: for  $|x| > r$ ,

$$E^s(x) = \sum_{n=1}^{\infty} \sum_{l=-n}^n \alpha_{nl}^{TE} S_n^{TE} E_{n,l}^{TE}(x; k_m) + \alpha_{nl}^{TM} S_n^{TM} E_{n,l}^{TM}(x; k_m),$$

where  $S_n^{TE}$  and  $S_n^{TM}$  are given by

$$\begin{aligned}S_n^{TE} &= \frac{\mu_c j_n(k_c r) \mathcal{J}_n(k_m r) - \mu_m j_n(k_m r) \mathcal{J}_n(k_c r)}{\mu_m \mathcal{J}_n(k_c r) h_n^{(1)}(k_m r) - \mu_c j_n(k_c r) \mathcal{H}(k_m r)}, \\ S_n^{TM} &= \frac{\varepsilon_c j_n(k_c r) \mathcal{J}_n(k_m r) - \varepsilon_m j_n(k_m r) \mathcal{J}_n(k_c r)}{\varepsilon_m \mathcal{J}_n(k_c r) h_n^{(1)}(k_m r) - \varepsilon_c j_n(k_c r) \mathcal{H}(k_m r)}.\end{aligned}$$

*Proof.* Let  $E^i = \tilde{E}_{n,l}^{TE}(x; k_m)$ . We look for a solution of the following form:

$$E = \begin{cases} a \tilde{E}_{n,l}^{TE}(x; k_c), & |x| < r \\ \tilde{E}_{n,l}^{TE}(x; k_m) + b E_{n,l}^{TE}(x; k_m), & |x| > r. \end{cases}$$

Then, from the boundary condition on  $\partial D$ , we easily see that

$$\begin{pmatrix} j_n(k_m r) \\ \frac{1}{\mu_m} \mathcal{J}_n(k_m r) \end{pmatrix} = \begin{pmatrix} j_n(k_c r) & -h_n^{(1)}(k_m r) \\ \frac{1}{\mu_c} \mathcal{J}_n(k_c r) & -\frac{1}{\mu_m} \mathcal{H}_n(k_m r) \end{pmatrix} \begin{pmatrix} a \\ b \end{pmatrix}. \quad (5.30)$$

Therefore, the coefficient  $a$  and  $b$  can be obtained as follows:

$$\begin{aligned} \begin{pmatrix} 1/a \\ b/a \end{pmatrix} &= \begin{pmatrix} j_n(k_m r) & h_n^{(1)}(k_m r) \\ \frac{1}{\mu_m} \mathcal{J}_n(k_m r) & \frac{1}{\mu_m} \mathcal{H}_n(k_m r) \end{pmatrix}^{-1} \begin{pmatrix} j_n(k_c r) & h_n^{(1)}(k_c r) \\ \frac{1}{\mu_c} \mathcal{J}_n(k_c r) & \frac{1}{\mu_c} \mathcal{H}_n(k_c r) \end{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \\ &= \frac{\mu_m k_m r}{i} \begin{pmatrix} \frac{1}{\mu_m} \mathcal{H}_n(k_m r) & -h_n^{(1)}(k_m r) \\ -\frac{1}{\mu_m} \mathcal{J}_n(k_m r) & j_n(k_m r) \end{pmatrix} \begin{pmatrix} j_n(k_c r) \\ \frac{1}{\mu_c} \mathcal{J}_n(k_c r) \end{pmatrix}, \\ &= -i k_m r \begin{pmatrix} \mathcal{H}_n(k_m r) j_n(k_c r) - \frac{\mu_m}{\mu_c} h_n^{(1)}(k_m r) \mathcal{J}_n(k_c r) \\ -\mathcal{J}_n(k_m r) j_n(k_c r) + \frac{\mu_m}{\mu_c} j_n(k_m r) \mathcal{J}_n(k_c r) \end{pmatrix}, \end{aligned} \quad (5.31)$$

where we have used the following Wronskian identity for the spherical Bessel function:

$$j_n(t) \mathcal{H}_n(t) - h_n^{(1)}(t) \mathcal{J}_n(t) = t \left( j_n(t) (h_n^{(1)})'(t) - j_n'(t) h_n^{(1)}(t) \right) = \frac{i}{t}.$$

Therefore, we immediately see that

$$b = \frac{\mu_c j_n(k_c r) \mathcal{J}_n(k_m r) - \mu_m j_n(k_m r) \mathcal{J}_n(k_c r)}{\mu_m \mathcal{J}_n(k_c r) h_n^{(1)}(k_m r) - \mu_c j_n(k_c r) \mathcal{H}_n(k_m r)}.$$

Now suppose that  $E^i = \tilde{E}_{n,l}^{TM}(x; k_m)$ . We look for a solution in the following form:

$$E = \begin{cases} c \tilde{E}_{n,l}^{TM}(x; k_c), & |x| < r, \\ \tilde{E}_{n,l}^{TM}(x; k_m) + d E_{n,l}^{TM}(x; k_m), & |x| > r. \end{cases}$$

Then, from the boundary conditions on  $|x| = r$ , we obtain

$$\begin{pmatrix} \frac{1}{\varepsilon_c} \mathcal{J}_n(k_c r) & \frac{1}{\varepsilon_c} \mathcal{H}_n(k_c r) \\ j_n(k_c r) & h_n^{(1)}(k_c r) \end{pmatrix} \begin{pmatrix} c \\ d \end{pmatrix} = \begin{pmatrix} \frac{1}{\varepsilon_m} \mathcal{J}_n(k_m r) & \frac{1}{\varepsilon_m} \mathcal{H}_n(k_m r) \\ j_n(k_m r) & h_n^{(1)}(k_m r) \end{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix}. \quad (5.32)$$

By solving (5.32), we get

$$d = \frac{\varepsilon_c j_n(k_c r) \mathcal{J}_n(k_m r) - \varepsilon_m j_n(k_m r) \mathcal{J}_n(k_c r)}{\varepsilon_m \mathcal{J}_n(k_c r) h_n^{(1)}(k_m r) - \varepsilon_c j_n(k_c r) \mathcal{H}_n(k_m r)}.$$

By the principle of superposition, the conclusion immediately follows. □

We also need the following lemma concerning the scattering amplitude  $A_\infty$ .

**Lemma 5.3.** *Suppose that the scattered electric field  $E^s$  is given by*

$$E^s(x) = \sum_{n=1}^{\infty} \sum_{l=-n}^n \beta_{nl}^{TE} E_{n,l}^{TE}(x; k_m) + \beta_{nl}^{TM} E_{n,l}^{TM}(x; k_m)$$

for  $\mathbb{R}^3 \setminus \overline{D}$ . Then the scattering amplitude  $A_\infty$  can be represented as follows:

$$A_\infty(\hat{x}) = \sum_{n=1}^{\infty} \sum_{l=-n}^n \frac{4\pi(-i)^n}{ik_m} \sqrt{n(n+1)} \left( \beta_{nl}^{TE} V_{n,l}(\hat{x}) + \sqrt{\frac{\mu_m}{\varepsilon_m}} \beta_{nl}^{TM} U_{n,l} \right).$$

*Proof.* It is well-known that

$$h_n^{(1)}(t) \sim \frac{1}{t} e^{it} e^{-i\frac{n+1}{2}\pi} \quad \text{as } t \rightarrow \infty,$$

and

$$(h_n^{(1)})'(t) \sim \frac{1}{t} e^{it} e^{-i\frac{n}{2}\pi} \quad \text{as } t \rightarrow \infty.$$

Then one can easily see that as  $|x| \rightarrow \infty$ ,

$$E_{n,m}^{TE}(x; k_m) \sim -\frac{e^{ik_m|x|}}{k_m|x|} e^{-i\frac{n+1}{2}\pi} \sqrt{n(n+1)} V_{n,l}(\hat{x})$$

and

$$E_{n,m}^{TM}(x; k_m) \sim -\frac{e^{ik_m|x|}}{k_m|x|} \sqrt{\frac{\mu_m}{\varepsilon_m}} e^{-i\frac{n+1}{2}\pi} \sqrt{n(n+1)} U_{n,l}(\hat{x}).$$

By applying these asymptotics to the series expansion of  $E^s$ , the conclusion follows. □

A plane wave can be represented as a series expansion. The following lemma is proved in [36].

**Lemma 5.4.** *Let  $E^i$  be a plane wave, that is,  $E^i(x) = p e^{ik_m d \cdot x}$  with  $d \in S$  and  $p \cdot d = 0$ . Then we have the following series representation for a plane wave as follows:*

$$E^i(x) = \sum_{n=1}^{\infty} \sum_{l=-n}^n \alpha_{nl}^{pw,TE} \tilde{E}_{n,l}^{TE}(x; k_m) + \alpha_{nl}^{pw,TM} \tilde{E}_{n,l}^{TM}(x; k_m),$$

where

$$\begin{cases} \alpha_{nl}^{pw,TE} = \frac{(-1)4\pi i^n}{\sqrt{n(n+1)}} i(V_{n,l}(d) \cdot p), \\ \alpha_{nl}^{pw,TM} = \frac{(-1)4\pi i^n}{\sqrt{n(n+1)}} \sqrt{\frac{\varepsilon_m}{\mu_m}} (U_{n,l}(d) \cdot p). \end{cases}$$

Now we are ready to compute the extinction cross-section  $Q^{ext}$ .



**Theorem 5.1.** Assume that  $E^i(x) = p e^{ik_m d \cdot x}$  with  $d \in S$  and  $p \cdot d = 0$ . Let  $D$  be a sphere with radius  $r$ . Then the extinction cross-section is given by

$$Q^{ext} = \sum_{n=1}^{\infty} \sum_{l=-n}^n \frac{(4\pi)^3}{k_m^2 |p|^2} \Im \left( (-1) S_n^{TE} (V_{n,l}(d) \cdot p)^2 + i S_n^{TM} (U_{n,l}(d) \cdot p)^2 \right).$$

Moreover, for small  $r > 0$ , we have

$$Q^{ext} = \sum_{l=-1}^1 \frac{(-1)(4\pi k_m r)^3}{k_m^2 |p|^2} \Im \left( i \frac{2}{3} \frac{\mu_c - \mu_m}{2\mu_m + \mu_c} (V_{1,l}(d) \cdot p)^2 + \frac{2}{3} \frac{\varepsilon_c - \varepsilon_m}{2\varepsilon_m + \varepsilon_c} (U_{1,l}(d) \cdot p)^2 \right) + O((k_m r)^4).$$

*Proof.* Let us first compute the scattering amplitude  $A_{\infty}$  when  $E^i$  is a plane wave. From

$$\begin{aligned} A_{\infty}(\hat{x}) &= \sum_{n=1}^{\infty} \sum_{l=-n}^n \frac{4\pi(-i)^n}{i k_m} \sqrt{n(n+1)} \\ &\quad \times \left( \alpha_{nl}^{pw,TE} S_n^{TE} V_{n,l}(\hat{x}) + \sqrt{\frac{\mu_m}{\varepsilon_m}} \alpha_{nl}^{pw,TM} S_n^{TM} U_{n,l} \right) \\ &= \sum_{n=1}^{\infty} \sum_{l=-n}^n \frac{(4\pi)^2}{k_m} \left( (-1) S_n^{TE} (V_{n,l}(d) \cdot p) V_{n,l} + i S_n^{TM} (U_{n,l}(d) \cdot p) U_{n,l} \right). \end{aligned}$$

Therefore, we have

$$\begin{aligned} Q^{ext} &= \frac{4\pi}{k_m} \Im \left[ \frac{p \cdot A_{\infty}(d)}{|p|^2} \right] \\ &= \sum_{n=1}^{\infty} \sum_{l=-n}^n \frac{(4\pi)^3}{k_m^2 |p|^2} \Im \left( (-1) S_n^{TE} (V_{n,l}(d) \cdot p)^2 + i S_n^{TM} (U_{n,l}(d) \cdot p)^2 \right). \end{aligned}$$

Now we assume that  $r \ll 1$ . By applying (5.20), one can easily see that

$$\begin{aligned} S_1^{TE} &= i \frac{2}{3} \frac{(\mu_c - \mu_m)(k_m r)^3}{2\mu_m + \mu_c} + O(r^4), \\ S_1^{TM} &= i \frac{2}{3} \frac{(\varepsilon_c - \varepsilon_m)(k_m r)^3}{2\varepsilon_m + \varepsilon_c} + O(r^4), \\ S_n^{TE}, S_n^{TM} &= O(r^4), \quad \text{for } n \geq 2. \end{aligned}$$

Therefore, we obtain, up to an error term of the order  $O(r^4)$ ,

$$Q^{ext} = \sum_{l=-1}^1 \frac{(-1)(4\pi)^3}{k_m^2 |p|^2} \Im \left( i \frac{2}{3} \frac{(\mu_c - \mu_m)(k_m r)^3}{2\mu_m + \mu_c} (V_{1,l}(d) \cdot p)^2 + \frac{2}{3} \frac{(\varepsilon_c - \varepsilon_m)(k_m r)^3}{2\varepsilon_m + \varepsilon_c} (U_{1,l}(d) \cdot p)^2 \right).$$

The proof is complete. □

## 6 Explicit computations for a spherical shell

### 6.1 Explicit representation of boundary integral operators

Let  $D_s$  and  $D_c$  be a spherical shell with radius  $r_s$  and  $r_c$  with  $r_s > r_c > 0$ . Let

$$(\varepsilon, \mu) = \begin{cases} (\varepsilon_m, \mu_m) & \text{in } D_c, \\ (\varepsilon_s, \mu_s) & \text{in } D_s \setminus \bar{D}_c, \\ (\varepsilon_m, \mu_m) & \text{in } \mathbb{R}^3 \setminus \bar{D}_s. \end{cases}$$

Let

$$\rho = \frac{r_c}{r_s}.$$

The solution to the transmission problem can be represented as follows

$$E(x) = \begin{cases} \mu_c \nabla \times \bar{\mathcal{S}}_{D_s}^{k_c}[\psi_s](x) + \nabla \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_c}[\phi_s](x) \\ \quad + \mu_c \nabla \times \bar{\mathcal{S}}_{D_c}^{k_c}[\psi_c](x) + \nabla \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_c}[\phi_c](x) & x \in D_c, \\ \mu_s \nabla \times \bar{\mathcal{S}}_{D_s}^{k_s}[\psi_s](x) + \nabla \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_s}[\phi_s](x) \\ \quad + \mu_s \nabla \times \bar{\mathcal{S}}_{D_c}^{k_s}[\psi_c](x) + \nabla \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_s}[\phi_c](x) & x \in D_s \setminus \bar{D}_c, \\ E^i + \mu_m \nabla \times \bar{\mathcal{S}}_{D_s}^{k_m}[\psi_s](x) + \nabla \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_m}[\phi_s](x) \\ \quad + \mu_m \nabla \times \bar{\mathcal{S}}_{D_c}^{k_m}[\psi_c](x) + \nabla \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_m}[\phi_c](x) & x \in \mathbb{R}^3 \setminus \bar{D}_s, \end{cases} \quad (6.1)$$

and

$$H(x) = -\frac{i}{\omega \mu_D} (\nabla \times E)(x) \quad x \in \mathbb{R}^3 \setminus \partial D, \quad (6.2)$$

where the pair  $(\psi_s, \phi_s, \psi_c, \phi_c) \in (H_T^{-\frac{1}{2}}(\text{div}, \partial D_s))^2 \times (H_T^{-\frac{1}{2}}(\text{div}, \partial D_c))^2$  is the unique solution to

$$W^{sh} \begin{pmatrix} \psi_s \\ \phi_s \\ \psi_c \\ \phi_c \end{pmatrix} := \begin{pmatrix} W_{11}^{sh} & W_{12}^{sh} \\ W_{21}^{sh} & W_{22}^{sh} \end{pmatrix} \begin{pmatrix} \psi_s \\ \phi_s \\ \psi_c \\ \phi_c \end{pmatrix} = \begin{pmatrix} \nu \times E^i \\ i\omega \nu \times H^i \\ 0 \\ 0 \end{pmatrix}$$

with

$$W_{11}^{sh} = \begin{pmatrix} \frac{\mu_s + \mu_m}{2} Id + \mu_s \mathcal{M}_{D_s}^{k_s} - \mu_m \mathcal{M}_{D_s}^{k_m} & \mathcal{L}_{D_s}^{k_s} - \mathcal{L}_{D_s}^{k_m} \\ \mathcal{L}_{D_s}^{k_s} - \mathcal{L}_{D_s}^{k_m} & \left( \frac{k_s^2}{2\mu_s} + \frac{k_m^2}{2\mu_m} \right) Id + \frac{k_s^2}{\mu_s} \mathcal{M}_{D_s}^{k_s} - \frac{k_m^2}{\mu_m} \mathcal{M}_{D_s}^{k_m} \end{pmatrix}, \quad (6.3)$$

$$W_{12}^{sh} = \begin{pmatrix} \mu_s \nu \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_s} - \mu_m \nu \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_m} & \nu \times \nabla \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_s} - \nu \times \nabla \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_m} \\ \nu \times \nabla \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_s} - \nu \times \nabla \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_m} & \frac{k_s^2}{\mu_s} \nu \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_s} - \frac{k_m^2}{\mu_m} \nu \times \nabla \times \bar{\mathcal{S}}_{D_c}^{k_m} \end{pmatrix} \Big|_{\partial D_s}, \quad (6.4)$$

$$W_{21}^{sh} = \begin{pmatrix} -\mu_c \nu \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_c} + \mu_s \nu \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_s} & -\nu \times \nabla \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_c} + \nu \times \nabla \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_s} \\ -\nu \times \nabla \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_c} + \nu \times \nabla \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_s} & -\frac{k_c^2}{\mu_c} \nu \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_c} + \frac{k_s^2}{\mu_s} \nu \times \nabla \times \bar{\mathcal{S}}_{D_s}^{k_s} \end{pmatrix} \Big|_{\partial D_c}, \quad (6.5)$$

$$W_{22}^{sh} = \begin{pmatrix} -\frac{\mu_c + \mu_s}{2} Id - \mu_c \mathcal{M}_{D_c}^{k_c} + \mu_s \mathcal{M}_{D_c}^{k_s} & -\mathcal{L}_{D_c}^{k_c} + \mathcal{L}_{D_c}^{k_s} \\ -\mathcal{L}_{D_c}^{k_c} + \mathcal{L}_{D_c}^{k_s} & -\left(\frac{k_c^2}{2\mu_c} + \frac{k_s^2}{2\mu_s}\right) Id - \frac{k_c^2}{\mu_c} \mathcal{M}_{D_c}^{k_c} + \frac{k_s^2}{\mu_s} \mathcal{M}_{D_c}^{k_s} \end{pmatrix}. \quad (6.6)$$

Note that  $W_{11}^{sh}$  and  $W_{22}^{sh}$  are similar to the operator in left-hand side of (3.4). In the previous section for the sphere case, we have already obtained the matrix representation of this operator and its asymptotic expansion.

By Lemma 5.1, we can represent  $\nu \times \nabla \times \vec{\mathcal{S}}_D^k|_{|x|=r'}$  and  $\nu \times \nabla \times \nabla \times \vec{\mathcal{S}}_D^k|_{|x|=r'}$  in a matrix form as follows (using  $U_{n,m}, V_{n,m}$  as basis):

(i) For  $r' > r$ ,

$$\nu \times \nabla \times \vec{\mathcal{S}}_D^k|_{|x|=r'} = \begin{pmatrix} (-ikr)\mathcal{J}_n(kr)h_n^{(1)}(kr') & 0 \\ 0 & ik\frac{r^2}{r'}j_n(kr)\mathcal{H}_n(kr') \end{pmatrix}, \quad (6.7)$$

$$\nu \times \nabla \times \nabla \times \vec{\mathcal{S}}_D^k|_{|x|=r'} = \begin{pmatrix} 0 & ik(kr)^2j_n(kr)h_n^{(1)}(kr') \\ -ik\frac{r}{r'}\mathcal{J}_n(kr)\mathcal{H}_n(kr') & 0 \end{pmatrix}; \quad (6.8)$$

(ii) For  $r' < r$ ,

$$\nu \times \nabla \times \vec{\mathcal{S}}_D^k|_{|x|=r'} = \begin{pmatrix} (-ikr)j_n(kr')\mathcal{H}_n(kr) & 0 \\ 0 & ik\frac{r^2}{r'}\mathcal{J}_n(kr')h_n^{(1)}(kr) \end{pmatrix}, \quad (6.9)$$

$$\nu \times \nabla \times \nabla \times \vec{\mathcal{S}}_D^k|_{|x|=r'} = \begin{pmatrix} 0 & ik(kr)^2j_n(kr')h_n^{(1)}(kr) \\ -ik\frac{r}{r'}\mathcal{J}_n(kr')\mathcal{H}_n(kr) & 0 \end{pmatrix}. \quad (6.10)$$

Using the above formulas, the matrix representation of the operators  $W_{12}^{sh}$  and  $W_{21}^{sh}$  can be easily obtained.

We now consider scaling of  $W^{sh}$ . First we need some definitions. Let  $D_s = z + r_s B_s$  where  $B_s$  contains the origin and  $|B_s| = O(1)$ . Let  $B_c$  be defined in a similar way. For any  $x \in \partial D_s$  (or  $\partial D_c$ ), let  $\tilde{x} = \frac{x-z}{r_s} \in \partial B_s$  (or  $\partial B_c$  with  $r_s$  replaced by  $r_c$ ) and define for each function  $f$  defined on  $\partial D_s$  (or  $\partial D_c$ ), a corresponding function defined on  $B$  as follows

$$\eta_s(f)(\tilde{x}) = f(z + r_s \tilde{x}), \quad \eta_c(f)(\tilde{x}) = f(z + r_c \tilde{x}). \quad (6.11)$$

Then, in a similar way to the sphere case, let us write

$$W_B^{sh}(r_s) \begin{pmatrix} \eta_s(\psi_s) \\ \omega \eta_s(\phi_s) \\ \eta_c(\psi_c) \\ \omega \eta_c(\phi_c) \end{pmatrix} = \begin{pmatrix} \frac{\eta(\nu \times E^i)}{\mu_m - \mu_s} \\ \frac{\eta(i\nu \times H^i)}{\varepsilon_m - \varepsilon_s} \\ 0 \\ 0 \end{pmatrix}.$$

Using  $(U_{n,m}, V_{n,m}, U_{n,m}, V_{n,m}) \times (U_{n,m}, V_{n,m}, U_{n,m}, V_{n,m})$  as basis, we can represent  $W_B^{sh}(r_s)$  in a  $8 \times 8$  matrix form in a subspace  $H_{n,m}(\partial B_s) \times H_{n,m}(\partial B_c)$ . Then, by using (5.21), their asymptotic expansion can also be obtained.

Here, the resulting asymptotics of the matrix  $W_B^{sh}$  are given as follows. Write

$$\mathcal{W}_B^{sh}(r_s) = \mathcal{W}_{B,0}^{sh} + r_s \mathcal{W}_{B,1}^{sh} + r_s^2 \mathcal{W}_{B,2}^{sh} + O(r_s^3), \quad (6.12)$$

where

$$\begin{aligned} \mathcal{W}_{B,0}^{sh} &= \begin{pmatrix} \Lambda_{\mu,\varepsilon} & \\ & \Lambda_{\mu,\varepsilon} \end{pmatrix} + \begin{pmatrix} P_{0,n} & Q_{0,n} \\ R_{0,n} & -P_{0,n} \end{pmatrix}, \\ \mathcal{W}_{B,1}^{sh} &= \begin{pmatrix} P_{1,n} & Q_{1,n} \\ R_{1,n} & -P_{1,n} \end{pmatrix}, \quad \mathcal{W}_{B,2}^{sh} = \begin{pmatrix} P_{2,n} & Q_{2,n} \\ R_{2,n} & -P_{2,n} \end{pmatrix}. \end{aligned} \quad (6.13)$$

Here, the matrix  $P_{j,n}$ ,  $Q_{j,n}$  and  $R_{j,n}$  are given by

$$\begin{aligned} \Lambda_{\mu,\varepsilon} &= \begin{pmatrix} \lambda_\mu & & & \\ & \lambda_\mu & & \\ & & \lambda_\varepsilon & \\ & & & \lambda_\varepsilon \end{pmatrix}, \quad P_{0,n} = \begin{pmatrix} p_n & & & \\ & -p_n & & \\ & & p_n & \\ & & & -p_n \end{pmatrix}, \\ Q_{0,n} &= \rho^2 \begin{pmatrix} g_n & & & \\ & f_n & & \\ & & g_n & \\ & & & f_n \end{pmatrix}, \quad R_{0,n} = \begin{pmatrix} f_n & & & \\ & g_n & & \\ & & f_n & \\ & & & g_n \end{pmatrix}, \\ P_{1,n} &= \omega \begin{pmatrix} & & C_\mu p_n \\ & C_\varepsilon p_n & C_\mu q_n \\ C_\varepsilon q_n & & \end{pmatrix}, \quad P_{2,n} = \omega^2 \begin{pmatrix} D_\mu r_n & & & \\ & D_\mu s_n & & \\ & & D_\varepsilon r_n & \\ & & & D_\varepsilon s_n \end{pmatrix}, \\ Q_{1,n} &= \omega \rho \begin{pmatrix} & & C_\mu \tilde{p}_n \\ & C_\varepsilon \tilde{p}_n & C_\mu \tilde{q}_n \\ C_\varepsilon \tilde{q}_n & & \end{pmatrix}, \quad Q_{2,n} = \omega^2 \rho \begin{pmatrix} D_\mu \tilde{r}_n & & & \\ & D_\mu \tilde{s}_n & & \\ & & D_\varepsilon \tilde{r}_n & \\ & & & D_\varepsilon \tilde{s}_n \end{pmatrix}, \\ R_{1,n} &= (-1)\omega \rho^{-1} \begin{pmatrix} & & C_\mu \tilde{p}_n \\ & C_\varepsilon \tilde{p}_n & C_\mu \tilde{q}_n \\ C_\varepsilon \tilde{q}_n & & \end{pmatrix}, \quad R_{2,n} = \omega^2 \rho^{-1} \begin{pmatrix} D_\mu \tilde{s}_n & & & \\ & -D_\mu \tilde{r}_n & & \\ & & D_\varepsilon \tilde{s}_n & \\ & & & -D_\varepsilon \tilde{r}_n \end{pmatrix}. \end{aligned}$$

Here,  $p_n, q_n, r_n, s_n$  are defined as (5.24) and  $\tilde{p}_n, \tilde{q}_n, \tilde{r}_n, \tilde{s}_n, D_\mu$  and  $D_\varepsilon$  are defined as follows:

$$f_n = \rho^n \frac{n}{2n+1}, \quad g_n = \rho^{n-1} \frac{n+1}{2n+1}, \quad (6.14)$$

$$\tilde{p}_n = \frac{1}{2n+1} \rho^{n+1}, \quad (6.15)$$

$$\tilde{q}_n = \frac{(n+1)(n-2)}{2(2n-1)(2n+1)} \rho^n - \frac{n(n+3)}{2(2n+1)(2n+3)} \rho^{n+2}, \quad (6.16)$$

$$\tilde{r}_n = -\frac{n+1}{2(2n-1)(2n+1)} \rho^n + \frac{(n+3)}{2(2n+1)(2n+3)} \rho^{n+2}, \quad (6.17)$$

$$\tilde{s}_n = -\frac{n-2}{2(2n-1)(2n+1)} \rho^{n+1} + \frac{n}{2(2n+1)(2n+3)} \rho^{n+3}, \quad (6.18)$$

and

$$D_\mu = \frac{\varepsilon_s \mu_s^2 - \varepsilon_m \mu_m^2}{\mu_m - \mu_s}, \quad D_\varepsilon = \frac{\varepsilon_s^2 \mu_s - \varepsilon_m^2 \mu_m}{\varepsilon_m - \varepsilon_s}. \quad (6.19)$$

## 6.2 Asymptotic behavior of the spectrum of $\mathcal{W}_B^{sh}(r_s)$

Let us define

$$\lambda_n^{sh} = \frac{1}{2(2n+1)} \sqrt{1 + 4n(n+1)\rho^{2n+1}}.$$

Note that  $\pm \lambda_n^{sh}$  are eigenvalues of the Neumann-Poincaré operator on the shell.

It turns out that the eigenvalues of  $W_{B,0}^{sh}$  are as follows

$$\lambda_\mu + \lambda_n^{sh}, \quad \lambda_\mu - \lambda_n^{sh}, \quad \lambda_\varepsilon + \lambda_n^{sh}, \quad \lambda_\varepsilon - \lambda_n^{sh},$$

for  $n = 0, 1, 2, \dots$ , and their multiplicities is 2. Their associated eigenfunctions are as follows:

$$\begin{aligned} \lambda_\mu + \lambda_n^{sh} &\longrightarrow E_1^0 := (\lambda_n^{sh} + p_n) \mathbf{e}_1 + f_n \mathbf{e}_5, & E_2^0 &:= (\lambda_n^{sh} - p_n) \mathbf{e}_2 + g_n \mathbf{e}_6, \\ \lambda_\mu - \lambda_n^{sh} &\longrightarrow E_3^0 := (-\lambda_n^{sh} + p_n) \mathbf{e}_1 + f_n \mathbf{e}_5, & E_4^0 &:= (-\lambda_n^{sh} - p_n) \mathbf{e}_2 + g_n \mathbf{e}_6, \\ \lambda_\varepsilon + \lambda_n^{sh} &\longrightarrow E_5^0 := (\lambda_n^{sh} + p_n) \mathbf{e}_3 + f_n \mathbf{e}_7, & E_6^0 &:= (\lambda_n^{sh} - p_n) \mathbf{e}_4 + g_n \mathbf{e}_8, \\ \lambda_\varepsilon - \lambda_n^{sh} &\longrightarrow E_7^0 := (-\lambda_n^{sh} + p_n) \mathbf{e}_3 + f_n \mathbf{e}_7, & E_8^0 &:= (-\lambda_n^{sh} - p_n) \mathbf{e}_4 + g_n \mathbf{e}_8, \end{aligned}$$

where  $\{\mathbf{e}_i\}_{i=1}^8$  is standard unit basis in  $\mathbb{R}^8$ .

To derive asymptotic expansions of the eigenvalues, we apply degenerate eigenvalue perturbation theory (since the multiplicity of each of these eigenvalues is 2). To state the result, we

need some definitions. Let

$$\begin{aligned}
T_{16,n} &= C_\varepsilon \frac{(L - p_n)a_{1,n} - b_{1,n}}{|E_1^0||E_6^0|}, & T_{18,n} &= C_\varepsilon \frac{(-L - p_n)a_{1,n} - b_{1,n}}{|E_1^0||E_8^0|}, \\
T_{25,n} &= C_\varepsilon \frac{(L + p_n)a_{2,n} - b_{2,n}}{|E_2^0||E_5^0|}, & T_{27,n} &= C_\varepsilon \frac{(-L + p_n)a_{2,n} - b_{2,n}}{|E_2^0||E_7^0|}, \\
T_{36,n} &= C_\varepsilon \frac{(L - p_n)a_{3,n} - b_{3,n}}{|E_3^0||E_6^0|}, & T_{38,n} &= C_\varepsilon \frac{(-L - p_n)a_{3,n} - b_{3,n}}{|E_3^0||E_8^0|}, \\
T_{45,n} &= C_\varepsilon \frac{(L + p_n)a_{4,n} - b_{4,n}}{|E_4^0||E_5^0|}, & T_{47,n} &= C_\varepsilon \frac{(-L + p_n)a_{4,n} - b_{4,n}}{|E_4^0||E_7^0|}, \\
T_{52,n} &= \frac{C_\mu}{C_\varepsilon} T_{16,n}, & T_{54,n} &= \frac{C_\mu}{C_\varepsilon} T_{18,n}, & T_{61,n} &= \frac{C_\mu}{C_\varepsilon} T_{25,n}, & T_{63,n} &= \frac{C_\mu}{C_\varepsilon} T_{27,n}, \\
T_{72,n} &= \frac{C_\mu}{C_\varepsilon} T_{36,n}, & T_{74,n} &= \frac{C_\mu}{C_\varepsilon} T_{38,n}, & T_{81,n} &= \frac{C_\mu}{C_\varepsilon} T_{45,n}, & T_{83,n} &= \frac{C_\mu}{C_\varepsilon} T_{47,n},
\end{aligned}$$

where

$$\begin{aligned}
a_{1,n} &= (\lambda_n^{sh} + p_n)q_n + \rho f_n \tilde{q}_n, \\
a_{2,n} &= (\lambda_n^{sh} - p_n)p_n + \rho g_n \tilde{p}_n, \\
a_{3,n} &= (-\lambda_n^{sh} + p_n)q_n + \rho f_n \tilde{q}_n, \\
a_{4,n} &= (-\lambda_n^{sh} - p_n)p_n + \rho g_n \tilde{p}_n,
\end{aligned}$$

and

$$\begin{aligned}
b_{1,n} &= f_n g_n q_n + \rho^{-1}(\lambda_n^{sh} + p_n)g_n \tilde{q}_n, \\
b_{2,n} &= f_n g_n p_n + \rho^{-1}(\lambda_n^{sh} - p_n)f_n \tilde{p}_n, \\
b_{3,n} &= f_n g_n q_n + \rho^{-1}(-\lambda_n^{sh} + p_n)g_n \tilde{q}_n, \\
b_{4,n} &= f_n g_n p_n + \rho^{-1}(-\lambda_n^{sh} - p_n)f_n \tilde{p}_n.
\end{aligned}$$

We also define

$$\begin{aligned}
K_{1,n} &= D_\mu \frac{(\lambda_n^{sh} + p_n)((\lambda_n^{sh} + p_n)r_n + \rho f_n \tilde{r}_n) + f_n((\lambda_n^{sh} + p_n)\rho^{-1} \tilde{s}_n - f_n r_n)}{|E_1^0|^2}, \\
K_{2,n} &= D_\mu \frac{g_n((-\lambda_n^{sh} + p_n)\rho^{-1} \tilde{r}_n - g_n s_n) + (\lambda_n^{sh} - p_n)((\lambda_n^{sh} - p_n)s_n + \rho g_n \tilde{s}_n)}{|E_2^0|^2}, \\
K_{3,n} &= D_\mu \frac{(-\lambda_n^{sh} + p_n)((-\lambda_n^{sh} + p_n)r_n + \rho f_n \tilde{r}_n) + f_n((-\lambda_n^{sh} + p_n)\rho^{-1} \tilde{s}_n - f_n r_n)}{|E_3^0|^2}, \\
K_{4,n} &= D_\mu \frac{g_n((\lambda_n^{sh} + p_n)\rho^{-1} \tilde{r}_n - g_n s_n) + (-\lambda_n^{sh} - p_n)((-\lambda_n^{sh} - p_n)s_n + \rho g_n \tilde{s}_n)}{|E_4^0|^2}, \\
K_{5,n} &= \frac{D_\varepsilon}{D_\mu} K_{1,n}, & K_{6,n} &= \frac{D_\varepsilon}{D_\mu} K_{2,n}, & K_{7,n} &= \frac{D_\varepsilon}{D_\mu} K_{3,n}, & K_{8,n} &= \frac{D_\varepsilon}{D_\mu} K_{4,n}.
\end{aligned}$$

Now we are ready to state the result. The followings are asymptotics of eigenvalues of  $W_B^{sh}(r_s)$

$$\begin{aligned}
& \lambda_\mu + \lambda_\varepsilon + (r_s \omega)^2 \left( \frac{T_{16,n} T_{61,n}}{\lambda_\mu - \lambda_\varepsilon} + \frac{T_{18,n} T_{81,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} + K_{1,n} \right) + O(r_s^3), \\
& \lambda_\mu + \lambda_\varepsilon + (r_s \omega)^2 \left( \frac{T_{16,n} T_{61,n}}{\lambda_\mu - \lambda_\varepsilon} + \frac{T_{18,n} T_{81,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} + K_{2,n} \right) + O(r_s^3), \\
& \lambda_\mu - \lambda_\varepsilon + (r_s \omega)^2 \left( \frac{T_{36,n} T_{63,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} + \frac{T_{38,n} T_{83,n}}{\lambda_\mu - \lambda_\varepsilon} + K_{3,n} \right) + O(r_s^3), \\
& \lambda_\mu - \lambda_\varepsilon + (r_s \omega)^2 \left( \frac{T_{36,n} T_{63,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} + \frac{T_{38,n} T_{83,n}}{\lambda_\mu - \lambda_\varepsilon} + K_{4,n} \right) + O(r_s^3), \\
& \lambda_\varepsilon + \lambda_\mu + (r_s \omega)^2 \left( \frac{T_{52,n} T_{25,n}}{\lambda_\varepsilon - \lambda_\mu} + \frac{T_{54,n} T_{45,n}}{\lambda_\varepsilon - \lambda_\mu + 2\lambda_n^{sh}} + K_{5,n} \right) + O(r_s^3), \\
& \lambda_\varepsilon + \lambda_\mu + (r_s \omega)^2 \left( \frac{T_{52,n} T_{25,n}}{\lambda_\varepsilon - \lambda_\mu} + \frac{T_{54,n} T_{45,n}}{\lambda_\varepsilon - \lambda_\mu + 2\lambda_n^{sh}} + K_{6,n} \right) + O(r_s^3), \\
& \lambda_\varepsilon - \lambda_\mu + (r_s \omega)^2 \left( \frac{T_{72,n} T_{27,n}}{\lambda_\varepsilon - \lambda_\mu - 2\lambda_n^{sh}} + \frac{T_{74,n} T_{47,n}}{\lambda_\varepsilon - \lambda_\mu} + K_{7,n} \right) + O(r_s^3), \\
& \lambda_\varepsilon - \lambda_\mu + (r_s \omega)^2 \left( \frac{T_{72,n} T_{27,n}}{\lambda_\varepsilon - \lambda_\mu - 2\lambda_n^{sh}} + \frac{T_{74,n} T_{47,n}}{\lambda_\varepsilon - \lambda_\mu} + K_{8,n} \right) + O(r_s^3).
\end{aligned}$$

We also have the following asymptotic expansions of the eigenfunctions:

$$\begin{aligned}
& E_1^0 + r_s \omega \left( \frac{T_{16,n}}{\lambda_\mu - \lambda_\varepsilon} E_6^0 + \frac{T_{18,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} E_8^0 \right) + O(r_s^2), \\
& E_2^0 + r_s \omega \left( \frac{T_{25,n}}{\lambda_\mu - \lambda_\varepsilon} E_5^0 + \frac{T_{27,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} E_7^0 \right) + O(r_s^2), \\
& E_3^0 + r_s \omega \left( \frac{T_{36,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} E_6^0 + \frac{T_{38,n}}{\lambda_\mu - \lambda_\varepsilon} E_8^0 \right) + O(r_s^2), \\
& E_4^0 + r_s \omega \left( \frac{T_{45,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} E_5^0 + \frac{T_{47,n}}{\lambda_\mu - \lambda_\varepsilon} E_7^0 \right) + O(r_s^2), \\
& E_5^0 + r_s \omega \left( \frac{T_{52,n}}{\lambda_\mu - \lambda_\varepsilon} E_2^0 + \frac{T_{54,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} E_4^0 \right) + O(r_s^2), \\
& E_6^0 + r_s \omega \left( \frac{T_{61,n}}{\lambda_\mu - \lambda_\varepsilon} E_1^0 + \frac{T_{63,n}}{\lambda_\mu - \lambda_\varepsilon + 2\lambda_n^{sh}} E_3^0 \right) + O(r_s^2), \\
& E_7^0 + r_s \omega \left( \frac{T_{72,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} E_2^0 + \frac{T_{74,n}}{\lambda_\mu - \lambda_\varepsilon} E_4^0 \right) + O(r_s^2), \\
& E_8^0 + r_s \omega \left( \frac{T_{81,n}}{\lambda_\mu - \lambda_\varepsilon - 2\lambda_n^{sh}} E_1^0 + \frac{T_{83,n}}{\lambda_\mu - \lambda_\varepsilon} E_3^0 \right) + O(r_s^2).
\end{aligned}$$

Interestingly, the first-order term (of order  $\delta$ ) is still zero in the asypmtotic expansions of the eigenvalues. This is due to the fact that degenerate eigenfunctions does not interact with each other.

## 7 Plasmonic resonances for the anisotropic problem

In this section, we consider the scattering problem of a time-harmonic wave  $u^i$ , incident on a plasmonic anisotropic nanoparticle. The homogeneous medium is characterized by electric permittivity  $\varepsilon_m$ , while the particle occupying a bounded and simply connected domain  $\Omega \Subset \mathbb{R}^3$  of class  $\mathcal{C}^{1,\alpha}$  for  $0 < \alpha < 1$  is characterized by electric anisotropic permittivity  $A$ . We consider  $A$  to be a positive-definite symmetric matrix.

In the quasi-static regime the problem can be modeled as follows

$$\begin{aligned} \nabla \cdot (\varepsilon_m Id \chi(\mathbb{R}^3 \setminus \bar{\Omega}) + A \chi(\Omega)) \nabla u &= 0, \\ |u - u^i| &= O(|x|^{-2}), \quad |x| \rightarrow +\infty, \end{aligned} \tag{7.1}$$

where  $\chi$  denotes the characteristic function and  $u^i$  is a harmonic function in  $\mathbb{R}^3$ .

We are interested in finding the plasmonic resonances for problem (7.1).

First, introduce the fundamental solution to the operator  $\nabla \cdot A \nabla$  in dimension three

$$G^A(x) = -\frac{1}{4\pi \sqrt{\det(A)} |A_* x|}$$

with  $A_* = \sqrt{A^{-1}}$ . From now on we will note  $G^A(x, y) := G^A(x - y)$ .

The single-layer potential associated with  $A$  is

$$\begin{aligned} \mathcal{S}_\Omega^A[\varphi] : H^{-\frac{1}{2}}(\partial\Omega) &\longrightarrow H^{\frac{1}{2}}(\partial\Omega) \\ \varphi &\longmapsto \mathcal{S}_\Omega^A[\varphi](x) = \int_{\partial\Omega} G^A(x, y) \varphi(y) d\sigma(y), \quad x \in \mathbb{R}^3. \end{aligned}$$

We can represent the unique solution [9] to (7.1) in the following form:

$$u(x) = \begin{cases} u^i + \mathcal{S}_\Omega[\psi], & x \in \mathbb{R}^3 \setminus \bar{\Omega}, \\ \mathcal{S}_\Omega^A[\phi], & x \in \Omega, \end{cases}$$

where  $(\psi, \phi) \in (H^{-\frac{1}{2}}(\partial\Omega))^2$  is the unique solution to the following system of integral equations on  $\partial\Omega$ :

$$\begin{cases} \mathcal{S}_\Omega[\psi] - \mathcal{S}_\Omega^A[\phi] &= -u^i, \\ \varepsilon_m \frac{\partial \mathcal{S}_\Omega[\psi]}{\partial \nu} \Big|_+ - \nu \cdot A \nabla \mathcal{S}_\Omega^A[\phi] \Big|_- &= -\varepsilon_m \frac{\partial u^i}{\partial \nu}. \end{cases} \tag{7.2}$$

**Lemma 7.1.** *The operator  $\mathcal{S}_\Omega^A : H^{-\frac{1}{2}}(\partial\Omega) \rightarrow H^{\frac{1}{2}}(\partial\Omega)$  is invertible. Moreover, we have the jump formula*

$$\nu \cdot A \nabla \mathcal{S}_\Omega^A \Big|_\pm = \pm \frac{1}{2} Id + (\mathcal{K}_\Omega^A)^*,$$

with

$$(\mathcal{K}_\Omega^A)^*[\varphi](x) = \int_{\partial\Omega} -\frac{(x - y, \nu(x))}{4\pi \sqrt{\det(A)} |A_*(x - y)|^3} \varphi(y) d\sigma(y).$$



*Proof.* Let  $\mathcal{T}_{A_*} \in \mathcal{L}(H^s(\partial\tilde{\Omega}), H^s(\partial\Omega))$  be such that  $\mathcal{T}_{A_*}[\varphi](x) = \varphi(A_*x)$  for  $\varphi \in H^s(\partial\tilde{\Omega})$  and  $\tilde{\Omega} = A_*\Omega$ . Let  $r_\nu \in \mathcal{L}(H^s(\partial\Omega), H^s(\partial\Omega))$  be such that  $r_\nu[\varphi](x) = |A_*^{-1}\nu(x)|\varphi(x)$ . It follows by the change of variables  $\tilde{y} = A_*y$  that  $d\sigma(\tilde{y}) = \det\sqrt{A_*}|A_*^{-1}\nu(y)|d\sigma(y)$ . Thus,

$$\mathcal{S}_\Omega^A = \mathcal{T}_{A_*}\mathcal{S}_{\tilde{\Omega}}\mathcal{T}_{A_*}^{-1}r_\nu^{-1},$$

and in particular  $\mathcal{S}_\Omega^A$  is invertible and its inverse  $(\mathcal{S}_\Omega^A)^{-1} = r_\nu\mathcal{T}_{A_*}\mathcal{S}_{\tilde{\Omega}}^{-1}\mathcal{T}_{A_*}^{-1}$ .

Note that, for  $x \in \partial\Omega$ ,

$$\tilde{\nu}(\tilde{x}) = \frac{A_*^{-1}\nu(x)}{|A_*^{-1}\nu(x)|},$$

where  $\tilde{\nu}(\tilde{x})$  is the outward normal to  $\partial\tilde{\Omega}$  at  $\tilde{x} = A_*x$ . We have

$$\begin{aligned} \nu \cdot A\nabla\mathcal{S}_\Omega^A \Big|_\pm &= \nu \cdot A\nabla_x \left( \mathcal{T}_{A_*}\mathcal{S}_{\tilde{\Omega}}\mathcal{T}_{A_*}^{-1}r_\nu^{-1} \right) \Big|_\pm \\ &= \nu \cdot AA_* \left( \mathcal{T}_{A_*}\nabla_{\tilde{x}}\mathcal{S}_{\tilde{\Omega}}\mathcal{T}_{A_*}^{-1}r_\nu^{-1} \right) \Big|_\pm \\ &= |A_*^{-1}\nu|\tilde{\nu} \cdot \left( \mathcal{T}_{A_*}\nabla_{\tilde{x}}\mathcal{S}_{\tilde{\Omega}}\mathcal{T}_{A_*}^{-1}r_\nu^{-1} \right) \Big|_\pm \\ &= \pm \frac{1}{2}Id + (r_\nu\mathcal{T}_{A_*})\mathcal{K}_{\tilde{\Omega}}^*(r_\nu\mathcal{T}_{A_*})^{-1}. \end{aligned} \tag{7.3}$$

The result follows from a change of variables in the expression of the operator  $(\mathcal{K}_\Omega^A)^* := (r_\nu\mathcal{T}_{A_*})\mathcal{K}_{\tilde{\Omega}}^*(r_\nu\mathcal{T}_{A_*})^{-1}$ .  $\square$

**Lemma 7.2.**  $\mathcal{S}_\Omega^A$  is negative definite for the duality pairing  $(\cdot, \cdot)_{-\frac{1}{2}, \frac{1}{2}}$  and we can define a new inner product

$$(u, v)_{\mathcal{H}_A^*} = -(u, \mathcal{S}_\Omega^A[v])_{-\frac{1}{2}, \frac{1}{2}},$$

which is equivalent to  $(\cdot, \cdot)_{-\frac{1}{2}, \frac{1}{2}}$ .

*Proof.* Let  $\varphi \in H^{-\frac{1}{2}}(\partial\Omega)$ . Using Lemma 7.1, we have

$$\varphi = \nu \cdot A\nabla\mathcal{S}_\Omega^A[\varphi] \Big|_+ - \nu \cdot A\nabla\mathcal{S}_\Omega^A[\varphi] \Big|_-.$$

Thus

$$\begin{aligned} \int_{\partial\Omega} \varphi(x)\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) &= \int_{\partial\Omega} \nu \cdot A\nabla\mathcal{S}_\Omega^A[\varphi] \Big|_+ (x)\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) - \int_{\partial\Omega} \nu \cdot A\nabla\mathcal{S}_\Omega^A[\varphi] \Big|_- (x)\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) \\ &= - \int_{\mathbb{R}^3 \setminus \tilde{\Omega}} \nabla\mathcal{S}_\Omega^A[\varphi](x) \cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) - \int_{\mathbb{R}^3 \setminus \tilde{\Omega}} \mathcal{S}_\Omega^A[\varphi](x) \nabla \cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) \\ &\quad - \int_{\Omega} \nabla\mathcal{S}_\Omega^A[\varphi](x) \cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) + \int_{\Omega} \mathcal{S}_\Omega^A[\varphi](x) \nabla \cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) \\ &= - \int_{\mathbb{R}^3} \nabla\mathcal{S}_\Omega^A[\varphi](x) \cdot A\nabla\mathcal{S}_\Omega^A[\varphi](x)d\sigma(x) \leq 0, \end{aligned}$$

where the equality is achieved if and only if  $\varphi = 0$ . Here we have used an integration by parts, the fact that  $\mathcal{S}_\Omega^A[\varphi](x) = O(|x|^{-1})$  as  $|x| \rightarrow \infty$ ,  $\nabla \cdot A \nabla \mathcal{S}_\Omega^A[\varphi](x) = 0$  for  $x \in \mathbb{R}^3 \setminus \partial\Omega$  and that  $A$  is positive-definite.

In the same manner, it is known that

$$\|\varphi\|_{\mathcal{H}^*}^2 = \int_{\partial\Omega} \varphi(x) \mathcal{S}_\Omega^A[\varphi](x) d\sigma(x) = - \int_{\mathbb{R}^3} \nabla \mathcal{S}_\Omega[|\varphi](x)|^2 d\sigma(x).$$

Since  $A$  is positive-definite we have

$$c\|\varphi\|_{\mathcal{H}^*}^2 \leq \int_{\partial\Omega} \varphi(x) \mathcal{S}_\Omega^A[\varphi](x) d\sigma(x) \leq C\|\varphi\|_{\mathcal{H}^*}^2,$$

for some constants  $c$  and  $C$ .

Using the fact that  $(\cdot, \cdot)_{\mathcal{H}^*}$  is equivalent to  $(\cdot, \cdot)_{-\frac{1}{2}, \frac{1}{2}}$ , we get the desired result.  $\square$

From (7.2) we have  $\phi = (\mathcal{S}_\Omega^A)^{-1}(\mathcal{S}_\Omega[\psi] + u^i)$ , whereas, by Lemma 7.1, the following equation holds for  $\psi$ :

$$\mathcal{Q}_A[\psi] = F \tag{7.4}$$

with

$$\mathcal{Q}_A = \frac{1}{2}(\varepsilon_m Id + (\mathcal{S}_\Omega^A)^{-1} \mathcal{S}_\Omega) + (\varepsilon_m \mathcal{K}_\Omega^* - (\mathcal{K}_\Omega^A)^* (\mathcal{S}_\Omega^A)^{-1} \mathcal{S}_\Omega), \tag{7.5}$$

and

$$F = -\varepsilon_m \frac{\partial u^i}{\partial \nu} + \nu \cdot A \nabla \mathcal{S}_\Omega^A[(\mathcal{S}_\Omega^A)^{-1} u^i] \Big|_-.$$

**Theorem 7.1.**  $\mathcal{Q}_A$  has a countable number of eigenvalues.

*Proof.* It is clear that  $(\mathcal{K}_\Omega^A)^* : H^{-\frac{1}{2}}(\partial\Omega) \rightarrow H^{-\frac{1}{2}}(\partial\Omega)$  is a compact operator. Hence,  $\varepsilon_m \mathcal{K}_\Omega^* - (\mathcal{K}_\Omega^A)^* (\mathcal{S}_\Omega^A)^{-1} \mathcal{S}_\Omega$  is compact as well. Therefore, only the invertibility of  $\frac{1}{2}(\varepsilon_m Id + (\mathcal{S}_\Omega^A)^{-1} \mathcal{S}_\Omega)$  needs to be proven.

Since  $\mathcal{S}_\Omega^A$  is invertible, the invertibility of  $\frac{1}{2}(\varepsilon_m Id + (\mathcal{S}_\Omega^A)^{-1} \mathcal{S}_\Omega)$  is equivalent to that of  $\varepsilon_m \mathcal{S}_\Omega^A + \mathcal{S}_\Omega$ . Consider now, the bilinear form, for  $(\varphi, \psi) \in (H^{-\frac{1}{2}}(\partial\Omega))^2$

$$B(\varphi, \psi) = -\varepsilon_m \int_{\partial\Omega} \varphi(x) \mathcal{S}_\Omega^A[\psi](x) d\sigma(x) - \int_{\partial\Omega} \varphi(x) \mathcal{S}_\Omega[\psi](x) d\sigma(x).$$

From Lemma 7.2, we have

$$B(\psi, \psi) \geq C\|\psi\|_{H^{-\frac{1}{2}}(\partial\Omega)},$$

for some constant  $C > 0$ .

It follows then, from the Lax-Milgram theorem that  $\varepsilon_m \mathcal{S}_\Omega^A + \mathcal{S}_\Omega$  is invertible in  $H^{-\frac{1}{2}}(\partial\Omega)$ , whence the result.  $\square$

Recall that the electromagnetic parameter of the problem,  $A$ , depends on the frequency,  $\omega$  of the incident field. Therefore the operator  $\mathcal{Q}_A$  is frequency dependent and we should write

$\mathcal{Q}_A(\omega)$ .

Following definition 3, we say that  $\omega$  is a plasmonic resonance if

$$|\text{eig}_j(\mathcal{Q}_A(\omega))| \ll 1 \quad \text{and is locally minimal for some } j \in \mathbb{N},$$

where  $\text{eig}_j(\mathcal{Q}_A(\omega))$  stands for the  $j$ -th eigenvalue of  $\mathcal{Q}_A(\omega)$ .

Equivalently, we can say that  $\omega$  is a plasmonic resonance if

$$\omega = \arg \max_{\omega} \|\mathcal{Q}_A^{-1}(\omega)\|_{\mathcal{L}(\mathcal{H}^*(\partial\Omega))}. \quad (7.6)$$

From now on, we suppose that  $A$  is an anisotropic perturbation of an isotropic parameter, i.e.,  $A = \varepsilon_c(Id + P)$ , with  $P$  being a symmetric matrix and  $\|P\| \ll 1$ .

**Lemma 7.3.** *Let  $A = \varepsilon_c(Id + \delta R)$ , with  $R$  being a symmetric matrix,  $\|R\| = O(1)$  and  $\delta \ll 1$ . Let  $\text{Tr}$  denote the trace of a matrix. Then, as  $\delta \rightarrow 0$ , we have the following asymptotic expansions:*

$$\begin{aligned} \mathcal{S}_\Omega^A &= \frac{1}{\varepsilon_c} (\mathcal{S}_\Omega + \delta \mathcal{S}_{\Omega,1} + o(\delta)), \\ (\mathcal{S}_\Omega^A)^{-1} &= \varepsilon_c (\mathcal{S}_\Omega^{-1} + \delta \mathcal{B}_{\Omega,1} + o(\delta)), \\ (\mathcal{K}_\Omega^A)^* &= \mathcal{K}_\Omega^* + \delta \mathcal{K}_{\Omega,1}^* + o(\delta) \end{aligned}$$

with

$$\begin{aligned} \mathcal{S}_{\Omega,1}[\varphi](x) &= -\frac{1}{2} \text{Tr}(R) \mathcal{S}_\Omega[\varphi](x) - \frac{1}{2} \int_{\partial\Omega} \frac{(R(x-y), x-y)}{4\pi|x-y|^3} \varphi(y) d\sigma(y), \\ \mathcal{B}_{\Omega,1} &= -\mathcal{S}_\Omega^{-1} \mathcal{S}_{\Omega,1} \mathcal{S}_\Omega^{-1}, \\ \mathcal{K}_{\Omega,1}^* &= -\frac{1}{2} \text{Tr}(R) \mathcal{K}_\Omega^*[\varphi](x) - \frac{3}{2} \int_{\partial\Omega} \frac{(R(x-y), x-y)(x-y, \nu(x))}{4\pi|x-y|^5} \varphi(y) d\sigma(y). \end{aligned}$$

*Proof.* Recall that for  $\delta$  small enough

$$\begin{aligned} \sqrt{(I + \delta R)^{-1}} &= Id - \frac{\delta}{2} R + O(\delta^2), \\ \det(I + \delta R) &= 1 + \delta \text{Tr}(R) + o(\delta), \\ (1 + \delta x + o(\delta))^s &= 1 + \delta s x + o(\delta), \quad s \in \mathbb{R}. \end{aligned}$$

The results follow then from asymptotic expansions of  $-\frac{1}{4\pi\sqrt{\det(A)}|A_*x|^\beta}$ ,  $\beta = 1, 3$  and the identity

$$(\mathcal{S}_\Omega^A)^{-1} = \varepsilon_c (Id + \delta \mathcal{S}_\Omega^{-1} \mathcal{S}_{\Omega,1} + o(\delta))^{-1} \mathcal{S}_\Omega^{-1}.$$

□

Plugging the expressions above into the expression of  $\mathcal{Q}_A$  we get the following result.

**Lemma 7.4.** *As  $\delta \rightarrow 0$ , the operator  $\mathcal{Q}_A$  has the following asymptotic expansion*

$$\mathcal{Q}_A = \mathcal{Q}_{A,0} + \delta \mathcal{Q}_{A,1} + o(\delta),$$

where

$$\begin{aligned}\mathcal{Q}_{A,0} &= \frac{\varepsilon_m + \varepsilon_c}{2} Id + (\varepsilon_m - \varepsilon_c) \mathcal{K}_\Omega^*, \\ \mathcal{Q}_{A,1} &= \varepsilon_c \left( \left( \frac{1}{2} Id - \mathcal{K}_\Omega^* \right) \mathcal{B}_{\Omega,1} \mathcal{S}_\Omega - \mathcal{K}_{\Omega,1}^* \right).\end{aligned}$$

We regard the operator  $\mathcal{Q}_A$  as a perturbation of  $\mathcal{Q}_{A,0}$ . As in section 3, we use the standard perturbation theory to derive the perturbed eigenvalues and eigenvectors in  $\mathcal{H}^*(\partial\Omega)$ .

Let  $(\lambda_j, \varphi_j)$  be the eigenvalue and normalized eigenfunction pairs of  $\mathcal{K}_\Omega^*$  in  $\mathcal{H}^*(\partial\Omega)$  and  $\tau_j$  the eigenvalues of  $\mathcal{Q}_{A,0}$ . We have  $\tau_j = \frac{\varepsilon_m + \varepsilon_c}{2} + (\varepsilon_m - \varepsilon_c) \lambda_j$ . For simplicity, we consider the case when  $\lambda_j$  is a simple eigenvalue of the operator  $\mathcal{K}_\Omega^*$ . Define

$$P_{j,l} = (\mathcal{Q}_{A,1}[\varphi_j], \varphi_l)_{\mathcal{H}^*}.$$

As  $\delta \rightarrow 0$ , the perturbed eigenvalue and eigenfunction have the following form:

$$\begin{aligned}\tau_j(\delta) &= \tau_j + \delta \tau_{j,1} + o(\delta), \\ \varphi_j(\delta) &= \varphi_j + \delta \varphi_{j,1} + o(\delta),\end{aligned}$$

where

$$\begin{aligned}\tau_{j,1} &= P_{jj}, \\ \varphi_{j,1} &= \sum_{l \neq j} \frac{P_{jl}}{(\varepsilon_m - \varepsilon_c)(\lambda_j - \lambda_l)} \varphi_l.\end{aligned}$$

## 8 A Maxwell-Garnett theory for plasmonic nanoparticles

In this subsection we derive effective properties of a system of plasmonic nanoparticles. To begin with, we consider a bounded and simply connected domain  $\Omega \Subset \mathbb{R}^3$  of class  $\mathcal{C}^{1,\alpha}$  for  $0 < \alpha < 1$ , filled with a composite material that consists of a matrix of constant electric permittivity  $\varepsilon_m$  and a set of periodically distributed plasmonic nanoparticles with (small) period  $\eta$  and electric permittivity  $\varepsilon_c$ .

Let  $Y = ]-1/2, 1/2[^3$  be the unit cell and denote  $\delta = \eta^\beta$  for  $\beta > 0$ . We set the (rescaled) periodic function

$$\gamma = \varepsilon_m \chi(Y \setminus \bar{D}) + \varepsilon_c \chi(D),$$

where  $D = \delta B$  with  $B \Subset \mathbb{R}^3$  being of class  $\mathcal{C}^{1,\alpha}$  and the volume of  $B$ ,  $|B|$ , is assumed to be equal to 1. Thus, the electric permittivity of the composite is given by the periodic function

$$\gamma_\eta(x) = \gamma(x/\eta),$$

which has period  $\eta$ . Now, consider the problem

$$\nabla \cdot \gamma_\eta \nabla u_\eta = 0 \quad \text{in } \Omega \tag{8.1}$$

with an appropriate boundary condition on  $\partial\Omega$ . Then, there exists a homogeneous, generally anisotropic, permittivity  $\gamma^*$ , such that the replacement, as  $\eta \rightarrow 0$ , of the original equation (8.1) by

$$\nabla \cdot \gamma^* \nabla u_0 = 0 \quad \text{in } \Omega$$

is a valid approximation in a certain sense. The coefficient  $\gamma^*$  is called an effective permittivity. It represents the overall macroscopic material property of the periodic composite made of plasmonic nanoparticles embedded in an isotropic matrix.

The (effective) matrix  $\gamma^* = (\gamma_{pq}^*)_{p,q=1,2,3}$  is defined by [9]

$$\gamma_{pq}^* = \int_Y \gamma(x) \nabla u_p(x) \cdot \nabla u_q(x) dx,$$

where  $u_p$ , for  $p = 1, 2, 3$ , is the unique solution to the cell problem

$$\begin{cases} \nabla \cdot \gamma \nabla u_p = 0 & \text{in } Y, \\ u_p - x_p & \text{periodic (in each direction) with period 1,} \\ \int_Y u_p(x) dx = 0. \end{cases} \quad (8.2)$$

Using Green's formula, we can rewrite  $\gamma^*$  in the following form:

$$\gamma_{pq}^* = \varepsilon_m \int_{\partial Y} u_q(x) \frac{\partial u_p}{\partial \nu}(x) d\sigma(x). \quad (8.3)$$

The matrix  $\gamma^*$  depends on  $\eta$  as a parameter and cannot be written explicitly.

The following lemmas are from [9].

**Lemma 8.1.** *For  $p = 1, 2, 3$ , problem (8.2) has a unique solution  $u_p$  of the form*

$$u_p(x) = x_p + C_p + \mathcal{S}_{D\#}(\lambda_\varepsilon Id - \mathcal{K}_{D\#}^*)^{-1}[\nu_p](x) \quad \text{in } Y,$$

where  $C_p$  is a constant,  $\nu_p$  is the  $p$ -component of the outward unit normal to  $\partial D$ ,  $\lambda_\varepsilon$  is defined by (3.8), and

$$\begin{aligned} \mathcal{S}_{D\#}[\varphi](x) &= \int_{\partial D} G_{\#}(x, y) \varphi(y) d\sigma(y), \\ \mathcal{K}_{D\#}^*[\varphi](x) &= \int_{\partial D} \frac{\partial G_{\#}(x, y)}{\partial \nu(x)} \varphi(y) d\sigma(y) \end{aligned}$$

with  $G_{\#}(x, y)$  being the periodic Green function defined by

$$G_{\#}(x, y) = - \sum_{n \in \mathbb{Z}^3 \setminus \{0\}} \frac{e^{i2\pi n \cdot (x-y)}}{4\pi^2 |n|^2}.$$

**Lemma 8.2.** *Let  $\mathcal{S}_{D\#}$  and  $\mathcal{K}_{D\#}^*$  be the operators defined as in Lemma 8.1. Then the following*

trace formula holds on  $\partial D$

$$(\pm \frac{1}{2}Id + \mathcal{K}_{D\sharp}^*)[\varphi] = \frac{\partial \mathcal{S}_{D\sharp}[\varphi]}{\partial \nu} \Big|_{\pm}.$$

For the sake of simplicity, for  $p = 1, 2, 3$ , we set

$$\phi_p(y) = (\lambda_\varepsilon Id - \mathcal{K}_{D\sharp}^*)^{-1}[\nu_p](y) \quad \text{for } y \text{ in } \partial D \quad (8.4)$$

Thus, from Lemma 8.1, we get

$$\gamma_{pq}^* = \varepsilon_m \int_{\partial Y} (y_q + C_q + \mathcal{S}_{D\sharp}[\phi_q](y)) \frac{\partial(y_p + \mathcal{S}_{D\sharp}[\phi_p](y))}{\partial \nu} d\sigma(y).$$

Because of the periodicity of  $\mathcal{S}_{D\sharp}[\phi_p]$ , we get

$$\gamma_{pq}^* = \varepsilon_m \left( \delta_{pq} + \int_{\partial Y} y_q \frac{\partial \mathcal{S}_{D\sharp}[\phi_p]}{\partial \nu}(y) d\sigma(y) \right). \quad (8.5)$$

In view of the periodicity of  $\mathcal{S}_{D\sharp}[\phi_p]$ , the divergence theorem applied on  $Y \setminus \bar{D}$  and Lemma 8.2 yields (see [9])

$$\int_{\partial Y} y_q \frac{\partial \mathcal{S}_{D\sharp}[\phi_p]}{\partial \nu}(y) = \int_{\partial D} y_q \phi_p(y) d\sigma(y).$$

Let

$$\psi_p(y) = \phi_p(\delta y) \quad \text{for } y \in \partial B.$$

Then, by (8.5), we obtain

$$\gamma^* = \varepsilon_m (Id + fP), \quad (8.6)$$

where  $f = |D| = \delta^3 (= \eta^{3\beta})$  is the volume fraction of  $D$  and  $P = (P_{pq})_{p,q=1,2,3}$  is given by

$$P_{pq} = \int_{\partial B} y_q \psi_p(y) d\sigma(y). \quad (8.7)$$

To proceed with the computation of  $P$  we will need the following Lemma [9].

**Lemma 8.3.** *There exists a smooth function  $R(x)$  in the unit cell  $Y$  such that*

$$G_\sharp(x, y) = -\frac{1}{4\pi|x-y|} + R(x-y).$$

Moreover, the Taylor expansion of  $R(x)$  at 0 is given by

$$R(x) = R(0) - \frac{1}{6}(x_1^2 + x_2^2 + x_3^2) + O(|x|^4).$$

Now we can prove the main result of this section, which shows the validity of the Maxwell-

Garnett theory uniformly with respect to the frequency under the assumptions that

$$f \ll \text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))^{3/5} \quad \text{and} \quad (Id - \delta^3 R_{\lambda_\varepsilon(\omega)}^{-1} T_0)^{-1} = O(1), \quad (8.8)$$

where  $R_{\lambda_\varepsilon(\omega)}^{-1}$  and  $T_0$  are to be defined and  $\text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_D^*))$  is the distance between  $\lambda_\varepsilon(\omega)$  and the spectrum of  $\mathcal{K}_B^*$ .

**Theorem 8.1.** *Assume that (8.8) holds. Then we have*

$$\gamma^* = \varepsilon_m (Id + fM(Id - \frac{f}{3}M)^{-1}) + O\left(\frac{f^{8/3}}{\text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))^2}\right) \quad (8.9)$$

uniformly in  $\omega$ . Here,  $M = M(\lambda_\varepsilon(\omega), B)$  is the polarization tensor (4.1) associated with  $B$  and  $\lambda_\varepsilon(\omega)$ .

*Proof.* In view of Lemma 8.3 and (8.4), we can write, for  $x \in \partial D$ ,

$$(\lambda_\varepsilon(\omega)Id - \mathcal{K}_D^*)[\phi_p](x) - \int_{\partial D} \frac{\partial R(x-y)}{\partial \nu(x)} \phi_p(y) d\sigma(y) = \nu_p(x),$$

which yields, for  $x \in \partial B$ ,

$$(\lambda_\varepsilon(\omega)Id - \mathcal{K}_B^*)[\psi_p](x) - \delta^2 \int_{\partial B} \frac{\partial R(\delta(x-y))}{\partial \nu(x)} \psi_p(y) d\sigma(y) = \nu_p(x).$$

By virtue of Lemma 8.3, we get

$$\nabla R(\delta(x-y)) = -\frac{\delta}{3}(x-y) + O(\delta^3)$$

uniformly in  $x, y \in \partial B$ . Since  $\int_{\partial B} \psi_p(y) d\sigma(y) = 0$ , we now have

$$(R_{\lambda_\varepsilon(\omega)} - \delta^3 T_0 + \delta^5 T_1)[\psi_p](x) = \nu_p(x),$$

and so

$$(Id - \delta^3 R_{\lambda_\varepsilon(\omega)}^{-1} T_0 + \delta^5 R_{\lambda_\varepsilon(\omega)}^{-1} T_1)[\psi_p](x) = R_{\lambda_\varepsilon(\omega)}^{-1}[\nu_p](x), \quad (8.10)$$

where

$$\begin{aligned} R_{\lambda_\varepsilon(\omega)}[\psi_p](x) &= (\lambda_\varepsilon(\omega)Id - \mathcal{K}_B^*)[\psi_p](x), \\ T_0[\psi_p](x) &= \frac{\nu(x)}{3} \cdot \int_{\partial B} y \psi_p(y) d\sigma(y), \\ \|T_1\|_{\mathcal{L}(\mathcal{H}^*(\partial B))} &= O(1). \end{aligned}$$

Since  $\mathcal{K}_B^*$  is a compact self-adjoint operator in  $\mathcal{H}^*(\partial B)$  it follows that [30]

$$\|(\lambda_\varepsilon(\omega)Id - \mathcal{K}_B^*)^{-1}\|_{\mathcal{L}(\mathcal{H}^*(\partial B))} \leq \frac{c}{\text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))} \quad (8.11)$$

for a constant  $c$ .

It is clear that  $T_0$  is a compact operator. From the fact that the imaginary part of  $R_{\lambda_\varepsilon(\omega)}$  is

nonzero, it follows that  $Id - \delta^3 R_{\lambda_\varepsilon(\omega)}^{-1} T_0$  is invertible. Under the assumption

$$(Id - \delta^3 R_{\lambda_\varepsilon(\omega)}^{-1} T_0)^{-1} = O(1),$$

$$\delta^5 \ll \text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*)).$$

and (8.10), (8.11) we get

$$\begin{aligned} \psi_p(x) &= (Id - \delta^3 R_{\lambda_\varepsilon(\omega)}^{-1} T_0 + \delta^5 R_{\lambda_\varepsilon(\omega)}^{-1} T_1)^{-1} R_{\lambda_\varepsilon(\omega)}^{-1} [\nu_p](x) \\ &= (Id - \delta^3 R_{\lambda_\varepsilon(\omega)}^{-1})^{-1} R_{\lambda_\varepsilon(\omega)}^{-1} [\nu_p](x) + O\left(\frac{\delta^5}{\text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))}\right). \end{aligned}$$

Therefore, we obtain and estimate for  $\psi_p$

$$\psi_p = O\left(\frac{1}{\text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))}\right).$$

Now we multiply (8.10) by  $y_q$  and integrate over  $\partial B$ . We can derive from the estimate of  $\psi_p$  that

$$P(Id - \frac{f}{3}M) = M + O\left(\frac{\delta^5}{\text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))^2}\right),$$

and therefore,

$$P = M(Id + \frac{f}{3}M)^{-1} + O\left(\frac{\delta^5}{\text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))^2}\right),$$

with  $P$  being defined by (8.7). Since  $f = \delta^3$  and

$$M = O\left(\frac{\delta^3}{\text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))}\right),$$

it follows from (8.6) that the Maxwell-Garnett formula (8.9) holds (uniformly in the frequency  $\omega$ ) under the assumption (8.8) on the volume fraction  $f$ .  $\square$

**Remark 8.1.** As a corollary of Theorem 8.1, we see that in the case when  $fM = O(1)$ , which is equivalent to the scale  $f = O\left(\text{dist}(\lambda_\varepsilon(\omega), \sigma(\mathcal{K}_B^*))\right)$ , the matrix  $fM(Id - \frac{f}{3}M)^{-1}$  may have a negative-definite symmetric real part. This implies that the effective medium is plasmonic as well as anisotropic.

**Remark 8.2.** It is worth emphasizing that Theorem 8.1 does not only prove the validity of the Maxwell-Garnett theory but it can also be used together with the results in section 7 in order to derive the plasmonic resonances of the effective medium made of a dilute system of arbitrary-shaped plasmonic nanoparticles, following (7.6)

$$\omega = \arg \max_{\omega} \|\mathcal{Q}_{\gamma^*}^{-1}(\omega)\|_{\mathcal{L}(\mathcal{H}^*(\partial\Omega))}.$$



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